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Deep Neural Networks Applied to Forecast ^{60}Co Soil-Plant Transfer Factor Values Using Pedological Parameters

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ABSTRACT

The transfer of radionuclides from soil to plants determines the extent of food contamination and, consequently, the risk of radioactive exposure. Soil-plant transfer factor (F*v*) is an essential parameter for dose calculation due to ingestion of contaminated food, expressed by the ratio of the mass activity concentration of the plant to that of the soil. Different soil types and plant species result in a large variability in F*v* values for a single radionuclide. According to the literature, the main soil properties that could influence ^{60}Co F*v* are: mineralogy, texture, cation exchange capacity (CEC), soil organic matter (SOM), pH and nutrients. In this work, Artificial Neural Networks (ANN) was applied to evaluate the possibility to forecast ^{60}Co F*v* values based on edaphological indicators. The literature review conducted to struture ^{60}Co F*v* learning sets for radish root and radish leaf, using pH, SOM and CEC as edaphological indicators. This work showed that only deep neural networks (DNN) architecture with regularization layers was able to predict ^{60}Co F*v* values, using the three selected indicators. It was demonstrated by higher correlation for the training set (r=0.9996; n=17), as well for the validation (r=0.9997; n=20), for radish root, both sets with very low mean square errors. This architecture applied for radish leaves also showed good performance, despite the lower data number (n=15). The better performance of DNN was associated with the complexity of interaction between the three pedological parameters available for training, the size of the learning set and the appropriate edaphological indicators for ^{60}Co.

Keywords: **Cobalt, Systematic Review, Soil-Plant Transfer Factor, Neural Network.**

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1. INTRODUCTION

Radionuclides transferred from soil to plant represent a potential risk from exposure to radioactivity of the population, since the ingestion of these contaminated foods is the main pathway for increasing internal radiation doses [1]. Among the parameters used in computational models, the soil-plant transfer factor (F*v*) is a crucial information for evaluating the radiological risk due to food ingestion. F*v* relates the radionuclide activity in the edible part of the plant (Bq kg^{-1}, dry matter), to the soil activity (Bq kg^{-1}, dry matter) [2]. Several factors can affect F*v*, among them: soil properties, the physical-chemical form of the radionuclide, plant phenology and physiology, and levels of technologies involved in agricultural production [3,4]. A radionuclide can have different F*v* values with up to five orders of magnitude in a plant (or group of plants), depending on the soil properties and the time elapsed after contamination, as already observed for ^{137}Cs in cereals [5,6]. Thus, these differences result in a wide dispersion of F*v* values for a given radionuclide and the use of generic values such as those proposed by the IAEA [7] may underestimate or overestimate the regional values. Indeed, it was reported that ^{137}Cs F*v* values determined in Brazilian soils for cereals can be two orders of magnitude higher than ^{137}Cs F*v* values determined in European soils [8,9].

This expansion of boundaries allowed the consolidation of some principles that explain the behavior of ^{137}Cs in the soil-plant system and the identification of some relevant parameters. Based on the consolidation of the degree of knowledge achieved and the avaliable information, the application of computational methods, such as Artificial Neural Network (ANN) has become a possibility to forecast the soil to plant F*v* values for ^{137}Cs without the need for assays covering the universe of soils and edible species for the purpose of radiological risk assessment due to food ingestion [8]. It was found that for ^{90}Sr, there were still gaps in the knowledge that did not allow such a adjusted modeling as using ANN with a simple architecture, as it was possible for ^{137}Cs [10]. Rosa [11] also applied the same tool to test the prediction of F*v* values for ^{131}I and in this case there was little F*v* values information in the literature for its generalization, but the adjustment was perfect for the data set studied.

These results shows that there is the possibility of predicting F*v* values for other radionuclides using ANNs, since its behavior in the soil-plant system be well known and as well their indicator soil properties, followed, of course, by a minimum data number to create a learning set.

In radioecology, there is a hypothesis that states that soils with high F*v* values in one type of crop will also result in high F*v* values in other crop, which would mean that F*v* measurements for crops group can be used to estimate the radiovulnerability of an ecosystem to a given radionuclide [12].

^{60}Co is one of the main activation products of nuclear facilities, with a relatively high half-life (5.1 years) [13] and is generated as a result of activation with neutrons of the stable isotope ^{59}Co, that exists in structural components of nuclear reactor vessels [14]. ^{60}Co enters the environment as low-activity radioactive waste, routinely released in liquid effluent to aquatic environments next to nuclear facilities [15]. Although these releases are controlled and follow specific standards, they contribute to the inventory of radionuclides in the ecosystem [16,17]. According to Colle, Debet, and Real [18], radiocobalt did not significantly contaminate terrestrial environments due to its absence in atmospheric effluents from power plants and in the *fallout* from nuclear weapons testing. Moreover, the radiocobalt released from the Chernobyl accident was restricted to its vicinity. Hamilton [15] corroborates this observation, stating that radiocobalt is detected near the area it was released, mainly near the coastal zone and estuaries. Furthermore, in the Fukushima accident, cobalt was not detected in releases to the atmosphere, however large amounts of ^{60}Co were released to the Pacific Ocean [19]. Contamination of soils with radiocobalt can occur via irrigation of agricultural areas or through groundwater or floodings rivers [20,21]. An example of flooding occurred in December 2003, when adverse weather conditions led to the breaching of the dikes, causing a large flood in the lower part of the heavily used for agriculture Rhone Valley by the waters of the Rhone River, which receive low-activity liquid effluents from a large number of European nuclear facilities. This episode resulted in the transfer of about 204 MBq of ^{60}Co, among other radionuclides, through almost 700000 tons of sediment over an area of 60 km^2 [22]. In addition, the soil can also be contaminated with ^{60}Co through radiological accidents, such as the one that occurred in Goiânia (1987), or through improper disposal of contaminated waste [23]. From a non-radiological point of view, non-radioactive isotopes of cobalt can enter the soil when there is release from natural conditions, which includes volcanic eruption, sea spray, burning, weathering and anthropogenic activities, such as cobalt refinery, cobalt mining and agricultural additives [24, 25, 26]. These activities can generate excess of stable cobalt, considered a micronutrient for some plants.

The objective of this work was to apply ANNs to evaluate the possibility of forecasting ^{60}Co F*v* values as a function of pedological parameters considered relevant to its transfer processes in the soil-plant system. The availability of F*v* values based on regional properties allows a differentiated management, faster and more efficient measures in terms of radioprotection. Predicting the transfer of radionuclides in the environment is of interest from an environmental radioprotection perspective.

2. METHODOLOGY

2.1. Artificial neural networks

Artificial neural networks were developed inspired by generalizations of mathematical models of the biological nervous system and they attempt to model the information processing capacity of the human nervous system [27]. The most common type of ANN used in the analysis of environmental samples is the basic architecture of Multilayer Perceptrons (MLP) [28]. In a simple mathematical model of a neuron, the effects of synapses are represented by the weights of the connections that modulate the effect of the associated input signals, and the nerve impulse is computed as the sum of the weight of the input signals, transformed by a transfer function. The learning ability of an artificial neuron is achieved by adjusting the weights according to the chosen learning algorithm [29]. In these networks, neurons are organized into layers, the input and output layers being called visible and the intermediate layers, if any, called hidden. The artificial neural network model used in this work is illustrated in Figure 1. In this model, the first layer, or input layer, are the soil properties and the number of neurons adopted for the input layer was defined according to the available attributes, in this case 3 neurons. The hidden layers have the function of learning the interactions among soil parameters and their weights (represented by narrows) to explain F*v* values. The third layer was composed by one neuron with linear activation function. The output layer, in this case, is the F*v*. The combination of weights, which minimizes the error function, is considered to be a solution for the learning problem [8]. Both first and second layers used the logistic function as its activation function.

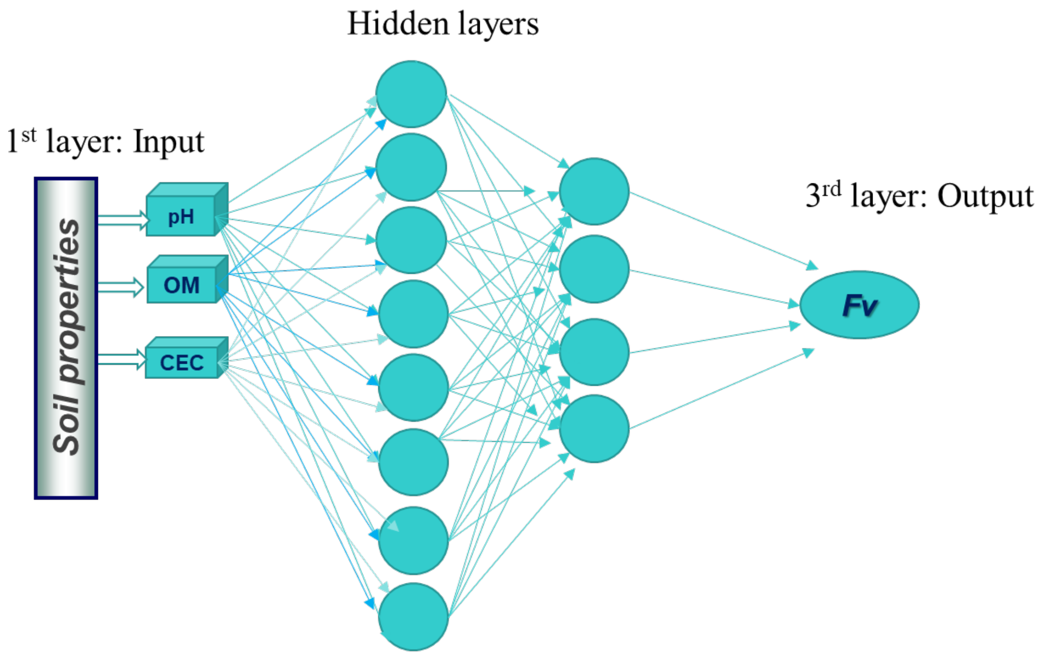

***Figure 1**: Artificial neural network model used in this work.*

The number of layers and neurons was varied, and for each architecture at least three random seeds were used. Preliminary testing pointed to the use of ReLU (Rectified Linear Unit) as activation functions. The Backpropagatiom algorithm was used for learning in which way the input parameters (soil properties) are related to F*v* values and, the Adam algorithm, was used as optimizer, with an initial learning rate (because the algorithm adapts it in training time) that overlapped with the commonly used stochastic gradient descent (SGD). The networks were developed in Python language, using the Tensorflow library.

In order for the generalization to be (as much as possible) evaluated during the model learning, the data set is separated into: i) Training set (training patterns), used for model fitting and this set should be representative of the data, containing most of them and; ii) Validation set (validation patterns), used to verify the achieved generalization for patterns that are not used in the creation of the model, also serving to guide the training stoppage, thus avoiding overfitting. Cross validation was used for the satisfaction criteria, where it is assumed that the best model found is the one that produces the minimum error on the validation set; and dropout was the regularization technique used to avoid overfitting, especially in larger networks (more layers and neurons) [30].

2.2. Systematic Literature Review for establish main pedological indicators for cobalt root uptake and Data Aquision

The objective of the systematic review was to survey the soil properties that most affected the behavior of cobalt in soils and to develop a learning set for artificial intelligence tools, using avaliable F*v* values for cobalt in the literature.

For the search process, the electronic databases consulted for the search of articles were: Google Scholar (https://scholar.google.com.br/), last viewed in 12/31/2021 and Periodic Capes (http://www.periodicos.capes.gov.br), last viewed in 12/31/2021. The keywords used in the search were "Plant-Soil relationships" and "Cobalt". The electronic search period was from 1986 to 2018. This periode was chosen due to the Chernobyl accident in 1986, which released large amounts of radionuclides into the environment, thus creating a field of interest in the subject. In these bases, the relevant documents were reviewed, considering the topic of interest (mainly biogeochemical processes related to the origin, fate, and behavior of cobalt in the soil-plant system) and presenting F*v* values. Additionally, it was performed manual searches for references cited in the expressive papers. The criteria for inclusion of F*v* values in the database were: a) F*v* values calculated through in situ studies and experimental studies provided that the methodological procedures were in accordance with the basis established by the International Union of Radioecologists [2]; b) F*v* values calculated manually through measurements of cobalt concentration in soils and plants, estimated by the obtained articles; c) F*v* values that contained at least one physicochemical property of the underlying soil; d) F*v* values in all plants of interest for human consumption. The search considered not only ^{60}Co, but also ^{57}Co and the stable isotope (^{59}Co).

Papers focused exclusively on foliar deposition, translocation and distribution of cobalt in plants, toxicity or health-related analysis, transfer to animals and to foods of animal origin, atmospheric processes, geological processes, and sorption on sediments in aquatic environments were excluded. The search process returned 2120 papers, from that, 1808 were in the exclusion criteria previously established and, from 312 papers reviewed, only 53 presented relevant F*v* data to this research.

The literature review, showed a gradual increase in the number of publications about cobalt. About 74% of the articles reviewed were published after 2008, but this crescent interesse cannot be attributed to the Fukushima accident (2011), as studies in aquatic environments were excluded from this analysis, despite the relevance of ^{60}Co releases in the Pacific Ocean [19]. One possible explanation is due to the industrial and technological advances in recent years, where cobalt releases from a variety of human activities are drawing the attention of researchers.

Reviewed articles with F*v* values by cobalt isotope are presented at Table 1. At this table is possible to observe that 36 articles presented 262 values for ^{59}Co F*v*. These articles mainly focus on the analysis of cobalt concentration levels in soils and plants. The 136 values for ^{60}Co F*v* presented in 17 articles show that the radiocobalt remains a radioecological concern. Furthermore, although ^{57}Co is an activation product in nuclear technologies, the restricted number of articles is probably due to its smaller half-life (270 days).

Table 1: Reviewed articles with F*v* values by cobalt isotope

Isotope	Number of F*v* values	Number of articles
^{59}Co	262	36
^{60}Co	136	17
^{57}Co	8	1

The main topics addressed in the 312 articles reviewed reinforce the predominance of studies that generate subsidies for the understanding of the behavior of cobalt in the soil-plant system: root uptake, phytoremediation, sorption mechanisms, human health and waste.

The journals with the most publications (more than 10) were: Environmental Monitoring and Assessment (16), Journal of Environmental Radioactivity (14), International Journal of Phytoremediation (13), Environmental Science and Pollution Research (13), Environmental Pollution (11) and Plant and Soil (11).

The literature review highlighted that pH, soil organic matter (SOM), mineralogy (mainly Mn and Fe oxides), texture, nutrients and cation exchange capacity (CEC) were the soil properties that showed some significant correlation with Co F*v* values. However, not all of those information is available simultaneously in one paper, associated with the F*v* value. Table 2 presents the soil properties that stood out as most relevant in the research for understanding the dynamics of cobalt in the soil-plant system, with their respective number of articles that addressed the topic.

Table 2: Soil properties referenced as relevant to understanding the dynamics of cobalt in the soil-plant system

Relevant soil properties	Number of articles that addressed the property
Mineralogy	68
pH	64
Organic matter	56
Texture	36
Nutrients	22
CEC	15

Moreover, some of these properties affect others, such as CEC which is a function of the mineralogy, the texture and the organic matter content of a soil, since it is defined as the sum of the negative charges on a solid soil particle's exchange sites and represents the ability of the soil to adsorb cations, and usually increases with higher levels of organic matter and clay minerals, as cation exchanges occurs on the surface of these components [31]. Or also pH, which modifies the charges of Fe and Mn oxides and indirectly can affect the CEC of a soil. The presence of nutrients can also affect the uptake of cobalt by plants and one of the possible explanations is that they compete with Co for the soil components's sorption sites [18]. In their work, Bartoly et al. [32] noted that in Brazilian Nitisols the transfer of ^{60}Co to plants is lower compared to Ferralsols and Acrisols and suggested that Nitisol is the most fertile soil of the three (higher values of CEC and higher levels of P, K and Ca), and thus may explain this disparity. There are indications that nutrients can impact the bioavailability of Co in soils, however there is no specific element that can be associated with Co and they are actually associated with the soil fertility level, which can indirectly be also evaluated by CEC.

Thus, these complex relationships between soil properties can, all together, overlap with each other, making the modeling to be arduous. Due to the lack of soil data associated to F*v* values, only three soil properties considered relevant and commonly measured in routine pedological analyses for agronomic purposes were selected to test the ability of the artificial neural network to predict ^{60}Co F*v* values. These are: pH, soil organic matter and CEC, this last parameter being understood as an indicator that integrates information about texture, mineralogy and soil fertility level, that means, per si, a very complex information.

In this review, 401 values were found for Co *Fv*, of which only 280 values contained any of the 3 soil properties indicative of the root absorption process associated with the respective F*v* value. However, these values referred to different cultures or parts of the same species, which resulted in incomplete learning groups, without the 3 input data of soil properties or with a really small amount of information. Thus, the database analysis returned only two datasets with enough information to establish a learning group for ANN training and validation: one for the radish root and other for the radish leaf.

3. RESULTS AND DISCUSSION

The present problem deal with the correlation between input patterns and their respective outputs (F*v*), which are provided to the network. A training pattern is then defined as a pair (X, Y) where X is the inputs and Y is the output. It is therefore a supervised learning, where a set of examples must be learned in order to obtain a model that generalizes, as best as possible, its behavior. Thus, a ANN was modeled for each problem. The identifications of these networks used in this work were: i) ANN-Root: neural network for predicting the transfer factor to radish root and ii) ANN-Leaf: neural network for predicting the transfer factor to radish leaf.

Thus, Tables 3 and 4 present, respectively, the database used for neural networks learning to forecast ^{60}Co F*v* for radish root (DB-Root) and for radish leaf (DB-Leaf) where suitable output (F*v*) and input values are shown, followed by the reference of each data. As the F*v* vary in orders of magnitude, they were submitted to logarithmic conversion to the base 10, in order to facilitate the ANN learning.

Table 3: Learning set for radish root (DB-Root) and reference of data

Pattern	***Fv***	***pH***	***OM***	***CEC***	*Reference*
1	1.40E-03	5.9	1	8	[1]
2	3.00E-03	6.3	23	8	[33]
3	3.00E-03	6.5	18	8	[33]
4	3.40E-03	5.4	3	23	[1]
5	3.80E-03	6.6	28	7	[33]
6	1.10E-02	4.8	4	20	[1]
7	1.10E-02	5.6	1	7	[1]
8	1.10E-02	5.7	42	18	[33]
9	1.70E-02	6.3	69	14	[33]
10	3.10E-02	5.8	55	17	[33]
11	3.58E-02	6.5	2	11	[23]
12	4.80E-02	5.7	74	17	[33]
13	5.85E-02	6.7	59	17	[33]
14	5.90E-02	6.2	23	7	[33]
15	6.40E-02	6.0	49	23	[33]
16	2.85E+00	5.4	1	2	[23]
17	8.27E-01	4.7	2	5	[23]
18	1.50E+00	4.5	23	6	[23]
19	1.16E+01	4.7	5	11	[23]
20	8.44E+00	4.9	4	37	[23]

Table 4: Learning set for radish leaf (DB-Leaf) and reference of data

Pattern	***Fv***	***pH***	***OM***	***CEC***	*Reference*
1	2.40E-03	6.3	23	8	[33]
2	2.70E-03	6.6	28	7	[33]
3	3.90E-03	6.5	18	8	[33]
4	8.60E-03	5.7	42	18	[33]
5	2.20E-02	5.8	55	17	[33]
6	2.20E-02	6.3	69	14	[33]
7	3.80E-02	6.7	59	17	[33]
8	5.40E-02	5.7	74	17	[33]
9	7.50E-02	6.2	23	7	[33]
10	7.60E-02	6.0	49	23	[33]
11	2.20E-01	5.4	1	2	[23]
12	5.02E-01	4.7	2	5	[23]
13	1.07E+00	4.5	23	6	[23]
14	2.19E+00	4.7	5	11	[23]
15	4.96E+00	4.9	4	37	[23]

Due to the small amount of examples obtained, the removal of outliers was avoided because, based on previous tests, it removed points known to be important in regions of the poorly represented data domain. After preliminary investigations it was observed that 10000 epochs would be sufficient for the convergence of the largest networks proposed in this paper. Since cross validation was used for the satisfaction criteria, this parameter could become oversized for the smaller networks.

3.1. Shallow Neural Net architecture

Because of the limited data available, a small number of patterns were selected for validation. They were chosen to not portray a representative part of the domain (because they are excluded from the learning set) and at the same time to express F*ν* values in different ranges. Therefore, from the available examples, three validation patterns were initially chosen (patterns 1, 9 and 12). Also due to the limited data, and aiming to avoid overfitting, the first architecture tested was a shallow network with only 1 hidden layer (ANN-Root-1). The amount of neurons in this layer was 120 neurons with ReLU activation function. Because it is a relatively small network, the dropout method was not applied to this network. The results of this ANN configuratiom originated Table 5, which displays the mean squared error (MSE) and the correlation coefficient (R2_Score) values for the training set and the validation set. The ANN-Root-1 also generated Figure 2, which presents a plot of real and predicted data for the training set and the validation set.

Table 5: Mean squared error (MSE) and the correlation coefficient (R2_Score) values for the training set and the validation set result for the ANN-Root-1 architeture

	Training	***Validation***
MSE	0.0124	0.0294
R2_Score	0.9916	0.9044

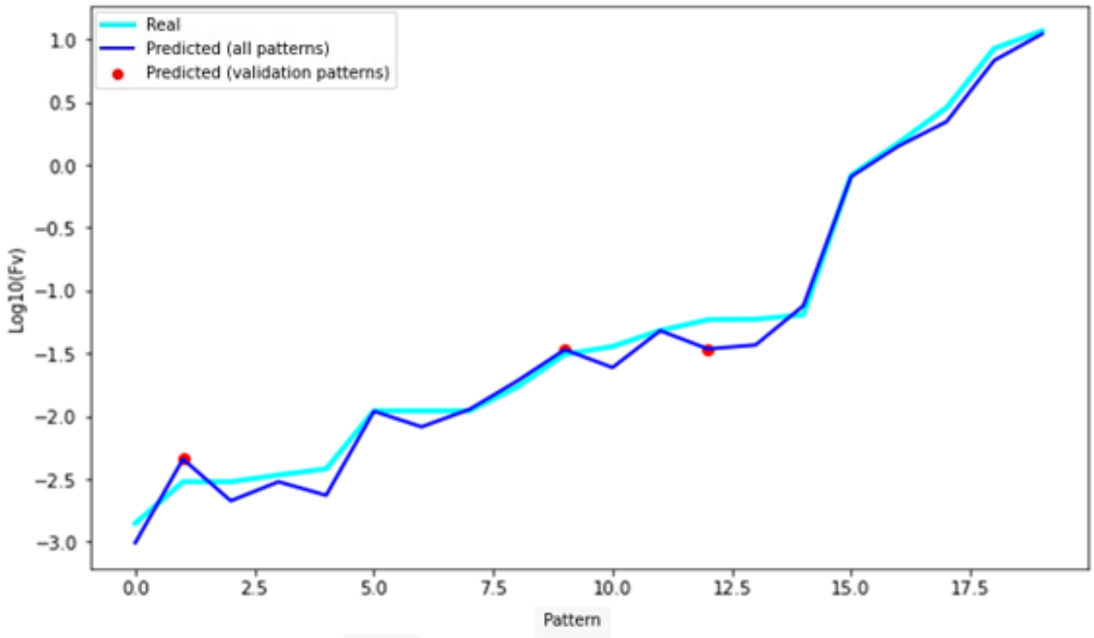

***Figure 2**: Plot of real and predicted data from ANN-Root-1*

It was observed that despite exhibiting an apparently reasonable correlation in the training set (r=0.9916; n=17), the graph in Figure 2 exhibits a considerable discrepancy from real data, mainly if we consider that data are reported in a logarithmic base. Also, the validation pattern differs from real data with lower correlation value (r=0.9044; n=20) and higher mean square errors (0.0294). Although the first tested architecture (ANN-Root-1) presented promising results, it was envisioned to improve the network by adding one more intermediate layer in a second architecture (ANN-Root-2), with more neurons and ReLU activation function. With the increase of the ANN size, the dropout method was implemented aiming to minimize overfitting. In the best ANN-Root-2 configuration, it was observed that it generated considerably better results than those obtained by ANN-Root-1. The predicted values for the validation patterns came closer to the real values, however a considerable discrepancy was still present.

3.2. Deep Neural Net architecture

Instead of increasing the number of neurons per layer as tested at the ANN-Root-2 architeture, it was decided to investigate deeper architectures (with more layers). Thus, the ANN-Root-3 was modeled with 3 intermediate layers with 120 neurons per layer, ReLU activation function and 2 dropout layers. The results obtained by the DNN-Root-3 architecture are presented in Table 6 and Figure 3. These results shows very high correlation value for the training set (r=0.9996;n=17), as well for the validation set (r=0.9997; n=20), both set with very low mean square errors: 0.0006 and 0.0001, respectivelly. Figure 3 exhibits a considerable consistence of predicted values for ^{60}Co F*v* with real data.

Table 6: Mean squared error (MSE) and the correlation coefficient (R2_Score) values for the training set and the validation set result for the DNN-Root-3 architeture

	Training	*Validation*
MSE	0.0006	0.0001
R2_Score	0.9996	0.9997

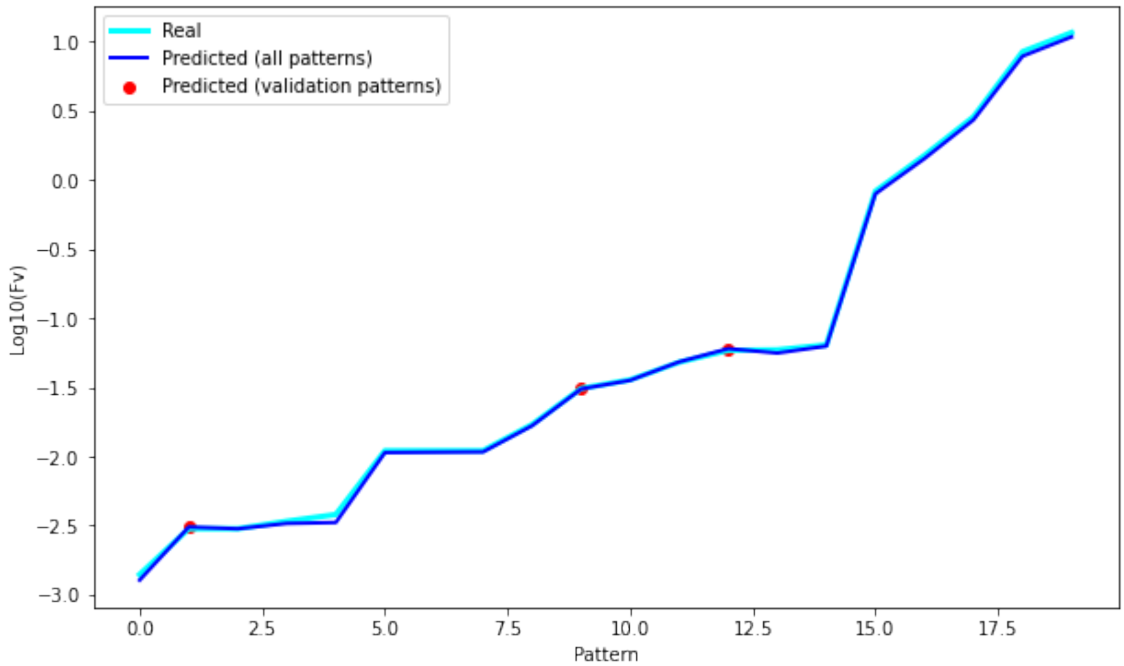

Figure 3: *Plot of real and predicted data from DNN-Root-3*

In this paper, the results of all experiments are not shown, only the results for the best architeture. The better results obtained with deeper networks is strongly linked to tree main factors: i) the use of rectified activation functions (ReLU), which minimizes the loss of gradient seen with the use of traditional functions, such as logistic and hyperbolic tangent; ii) the use of dropout regularization, which efficiently minimizes overfitting, and iii) CEC is a parameter that integrates others soils properties not discriminated as indicator in this study, due to the absence of data in the literature reported with F*v*, but also relevant to understand the behaviour of Co in soil: mineralogy, texture and soil fertility level. In brief, architetures with more neurons and more layers are more able to reach a better understanding of these relationship. That explains why a shalow ANN was able to achieve good results in predicting ^{137}Cs F*v* values: all the soil informations related to the behavior of ^{137}Cs in the soil-plant system were done as input layer, because they were easily available: exchangeable K, pH, OM and CEC [8].

Considering that the DNN architecture used for the radish root had excellent results, the same architecture was used for the validation of the DNN for the radish leaf, remembering that the learning set for the radish leaf is smaller than that for the root. The results of this ANN are presented in Table 7 and Figure 4.

Table 7: Mean squared error (MSE) and the correlation coefficient (R2_Score) values for the training set and the validation set result for the DNN-Leaf-1 architeture

	Training	***Validation***
MSE	0.0103	0.0002
R2_Score	0.9894	0.9997

It was observed that the results from DNN-Leaf-1 were also satisfactory, with high correlation coefficient for the validation set (r=0.9997) and low very low mean square errors (table 7). The training set also showed a good correlation coefficient (r=0.9894; n=12), although to a lesser extent than the DNN-Root-3. This was expected, due to the smaller number of patterns (fewer F*v* values) presented to this DNN compared to the DNN-Root-3. The DNN-Leaf-1 reinforces the power of this architeture for a set containing few data.

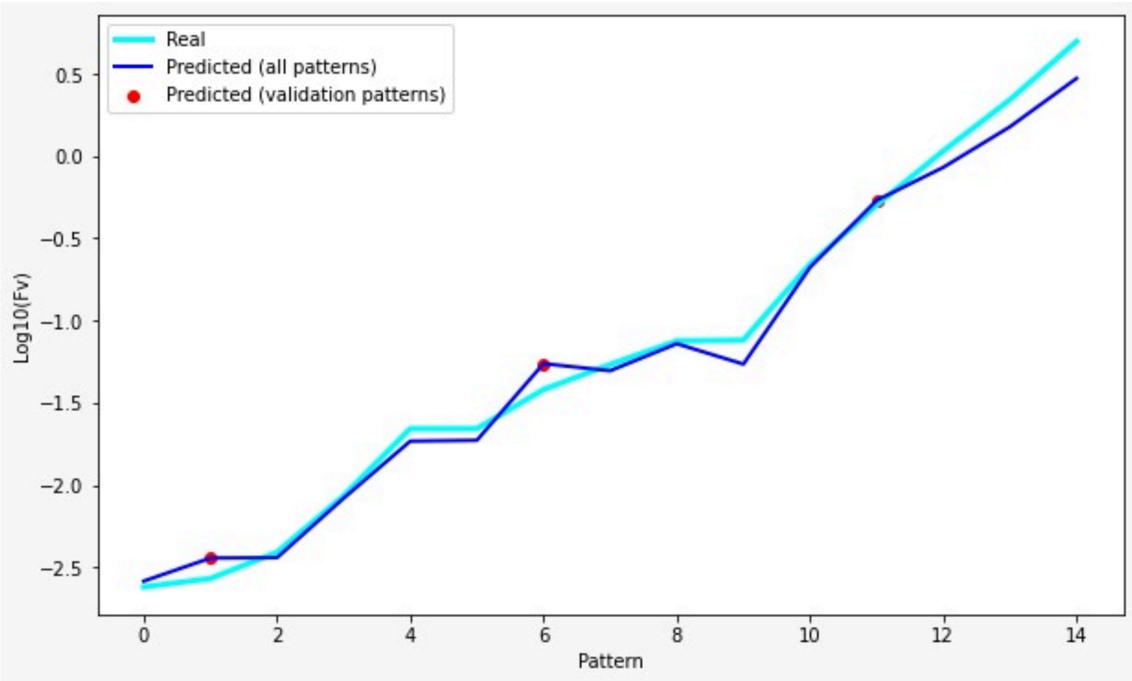

***Figure 4**: Plot of real and predicted data from ANN-Leaf-1*

DNNs seens to be a powerful tool for forecasting of ^{60}Co F*v* values provided that information from the selected pedological parameters is present: pH, organic matter content and CEC.

4. CONCLUSIONS

The literature review pointed out that mineralogy, soil organic matter, texture, pH, CEC and soil nutrients are the soil properties that best explain the behavior of ^{60}Co in the soil-plant system and therefore, are able to explain the F*v* values for this radionuclide. Initially, the indicators highlighted by this work, for the prediction of ^{60}Co F*v* values were pH, soil organic matter and CEC.

The use of a traditional ANN architecture showed a high correlation for training set ($r = 0.9916$; $n = 17$) but lower for validation set ($r=0.9044$; $n=20$) for radish root. This work showed that only deep neural networks (DNN) with regularization layers (dropout) were able to predict ^{60}Co F*v* values, using the information of the three selected edaphological indicators. It was demonstrated by higher correlation for training set (r=0.9996;n=17), as well for the validation (r=0.9997; n=20), for radish root, both set with very low mean square errors: 0.0006 and 0.0001, respectivelly. This architecture applied for radish leaves also showed a good performance, despite of the lower data number (n=15). The better performance of DNN, to the detriment of the superficial ANN, was associated with the complexity of the research space, that is, the complexity of interaction between the three pedological parameters available for training and the size of the learning set, however, the size of these sets does not seem to be a major limitation, although the prediction can always be improved with more information from values obtained in soils from different climates.

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