



# Prediction and Comparison of Nonlinear Mathematical Models for the Biodegradation of Two Herbicides Under the Effect of Manure in Soils

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<sup>14</sup>C<sub>2</sub>

## ABSTRACT

The study was for the comparison and to know the choice of Models of appreciation of the mineralization of the two herbicides under the effect of two manures (cattle and sheep) in two agricultural soils of different textures. During this work, we used two types of manure, cattle F1 and sheep F2 with two doses. The application of respirometry for monitoring biological activity has been conducted in the laboratory. The treatments were measured for carbon-labeled herbicides released (<sup>14</sup>CO<sub>2</sub>) after 1, 3, 7, 14, 28, 42, 60, 90, 120 and 150 days of incubation. Non-linear mathematical models have been developed for the study of the kinetics of the mineralization of herbicides under the effect of manures. The selection criteria for these fit models are R<sup>2</sup> and RMCE. The comparison of six models stated to choose the single-compartmental model to a first-order ascending exponential that best fits the experimental data. These models show a strong positive correlation between labeled carbon and the biodegradation time of herbicides, especially in clay-textured soil.

## INTRODUCTION

The repeated actions of the use of phytosanitary products intended to protect crops, over time lead to the sterilization of soils and consequently, the pollution of groundwater (Pimentel 1995, Auterives & Baran 2015). To do this, manure is added to degrade the herbicides into non-toxic substances to stimulate the native microflora (Entry & Emmingham 1995, Topp et al 1996, Savadogo et al 2008, Mansooreh 2013, Olu-arotiowa & Agarry 2019).

Our objective is to make mathematical modeling to predict the effect of manure spreading on the biodegradation of two herbicides in soils. Simulation of the results based on laboratory measurements of the labeled C of the herbicide.

To study the kinetics of carbon mineralization (<sup>14</sup>C<sub>Herbicide</sub>) of the herbicide glyphosate and 2,4-D, non-linear regression models were used. These are the zero-order models, then the first-order mono-compartmental exponential models, the two-compartmental double-exponential and two-compartmental exponential and linear models, and finally, the Hyperbolic model. The latter is compared for monitoring and selecting the adapted model of the kinetics of the mineralization of the herbicides. The parameters of choice are R<sup>2</sup> and RMCE.

## MATERIALS AND METHODS

### Sampling

Sampling consisted of taking two samples of agricultural soil of different textures from a depth of 30 cm. These samples were subjected to granulometry and physico-chemical analysis (Table 1).

The study of the soil analysis results recorded in the Tables (Table 1), reveals that soil 1 has a clay-sandy texture (G.E.P.P.A texture triangle) (Baize 1995), a slightly alkaline pH (7.54), a very high Electrical Conductivity (EC). A very low scale rate and the retention capacity is 32.54 % (a little high). The cation exchange capacity is 72 meq/100 g of soil. Organic Carbon is 2.13% and this gives the high organic matter rate of 3.66%, soil rich in humus. But the N c content is average and reaches 0.18% (Table 1) which gives a C/N ratio is 11.83 and this indicates a fairly good biological activity.

The soil analysis results reported in Table 1 show that soil 2 has a sandy texture (Baize 1995), a low alkaline pH (7.65), and an electrical conductivity (EC) of 50 μS/cm. The rate of limestone is very low (traces), the retention capacity

is 25.25% and the cationic exchange capacity is 16.65 meq/100g soil. Carbon C is 0.8% giving the low organic matter level of 1.38% (Table 2), but the N content is (0.08%) very low (Table 1). The C/N ratio is 10 and this indicates healthy soil and a good activity of the telluric microflora, that is the well decomposed organic matter, which means the stable soil humus.

It is inferred that the soil C content varies from 0.8% to 3.66%. The N content varies from 0.08 % to 0.18 %. The soils with the highest C and N content are clayey, and the C and N content of the soils is correlated with the clay content. The soil is low in clay (sandy), and has low C and N content.

### Choice of Herbicides and Their Doses

The two herbicides most commonly used in the study area are Glyphosate ( $C_3H_8NO_5P$ ) and 2,4-D ( $C_8H_6Cl_2O_3$ ). The agronomic dose (single) (D1) of 2.5  $\mu g$  for glyphosate and 2.1  $\mu g$  for 2,4-D was used.

### Manure Supply

We used two types of organic amendments. These are cattle manure (F1) and sheep manure (F2) of one year of age. The physico-chemical characteristics of the two manures are given in Table 2. The rate of the amendment is based on the organic matter content of the soil, which will be 5% for clay

Table 1: Texture and physico-chemical characteristics of the soils studied.

Physico-chemical characteristics	Unit of measure	Soil (S1)		Soil (S2)
Granulometry	%	sand	35	78
		Clay	15	10
		silt	40	12
Texture classes	Textured triangle (G.E.P.P.A)	Sandy-clays		Sandy
pH	-	7.54		7.65
Electrical conductivity CE	$\mu S/cm$	99.00		68
Water retention capacity CR	%	32.54		25.25
total limestone	%	0		0
$K^+$	ppm	173.10		57
$Na^+$	meq/100g	0.35		0.1
$Mg^{+2}$	meq/100g	1.84		0.32
$Ca^{+2}$	meq/100g	17.82		5.87
CEC	meq/100g	72		16.55
P (Olsen)	ppm	8.30		1.23
PT	ppm	175		28.22
NT	ppm	1800		800
C	%	2.13		0.8
OM	%	3.66		1.376
C/N	-	11.83		10

Table 2: Physico-chemical characteristics of the two manures.

Characteristics	Units	Cattle manure F1	Sheep manure F2
		F1D1	F2D1
Dry matter	%	28.4	29.3
$C_{org}$		13.43	14.65
N	g/kg	6.2	8.6
$P_2O_5$		1.41	1.8
$K_2O$		5.93	11.03
pH	-	7.9	8.3
C/N		21.66	17.03

soil and 15% for sandy soil. Analytical results for cattle and sheep manure are presented in Table 2.

The pH values balance between slightly basic to basic levels with pH 7.9 to 8.3 respectively for cattle manure (F1) and sheep manure (F2), a dry matter rate of 28.4% and 29.3% respectively for Cattle manure F1 and Sheep manure F2, which tells us about the pasty nature of these two types of manure. These two manures are moderately rich in total nitrogen with a content of 6.3 g/kg for F1 and 8.6 g/kg for F2. Levels 1.41 g/kg for F1 and 1.8 g/kg for F2 are considered as low P<sub>2</sub>O<sub>5</sub>. The organic fertilization guide GFOR in Chabali er et al. (2006) indicates that cattle manure is average (5.93 g/kg) and sheep manure is rich (11.03 g/kg) for K<sub>2</sub>O.

The C/N ratio, which indicates the rate of decomposition of organic matter, is thus deduced. Indeed, F1 to C/N slightly high (21.66) will cause slow mineralization or immobilization of nitrogen by terrestrial microorganisms because of its low nitrogen content. However, the other has a C/N of 17.03. According to Chabali er et al. (2006) manure F2, once brought to the ground, will quickly mineralize by supplying a lot of mineral nitrogen.

**Preparation of the Samples of the Experimental Device and the Determination of the 14C of the Herbicides**

The treatments for experimenting with the 14C mineralization of the two herbicides are:

- S1+H1, S1+H1+F1, S1+H1+F2
- S1+H2, S1+H2+F1, S1+H2+F2
- S2+H1, S2+H1+F1, S2+H1+F2
- S2+H2, S2+H2+F1, S2+H2+F2

The radiorespirometric device used consists of a one-liter jar, in which is placed a 250 mL glass jar (cup) containing 10 g of dry weight equivalent soil, a 20 mL liquid scintillation vial containing 5 mL of sodium hydroxide (NaOH 0.2 N) which traps the <sup>14</sup>CO<sub>2</sub> released during incubation at 28°C in the dark in flasks hermetically sealed with a rubber seal, is replaced regularly (after 1, 3, 7, 14, 21, 28, and 42, 60, 90, 120 and 150 days of incubation) and a 20 ml liquid scintillation vial containing 10 mL of distilled water to

saturate the atmosphere of the jar. Finally, the determination of radioactivity is carried out using a liquid scintillation meter (Soulas 1993).

For each soil type, three radiorespirometric devices containing the soil treated with a glyphosate solution (2.5 µg Glyphosate and 2000 Bq Glyphosate <sup>14</sup>C per sample), and three radiorespirometric devices containing the soil treated with a 2,4-D solution (2.1 µg of 2,4-D and 2000 Bq of <sup>14</sup>C-2,4-D per sample) were prepared. <sup>14</sup>C-glyphosate and <sup>14</sup>C-2,4-D were determined by the regional nuclear center of Algiers.

**Statistical Treatment of Results**

Nonlinear regression (NLR) is a method for determining a nonlinear model of the relationship between the dependent variable and a group of independent variables. This approach was used to model and simulate the carbon mineralization kinetics of the two herbicides, according to XLSTAT software. Six models were applied to estimate the mineralization potential of the two herbicides (Table 3). These are time-based mathematical models that use experimental results of herbicide biodegradation kinetics under the effect of both manures over a long incubation period (90 days).

Where,

Cm: represents the amount of carbon mineralized at time t;

Co: the quantity of potentially mineralizable carbon;

k: as mineralization rate constant;

t: represents the time.

In this study, we will deal with the comparative aspect of nonlinear adjustment models.

**RESULTS AND DISCUSSION**

**Comparisons and Selection of Models for Assessing Herbicide Mineralization by Non-Linear Regression**

The results obtained in standardized laboratory conditions (temperature 28°C, soil humidity 2/3 CR) reveal, a favorable

Table 3: The mathematical models used.

Model	Equation	Reference
M1: The single-compartment model (first-order exponential)	$Cm = Co*(1 - exp^{-k*t})$	Murwira et al. (1988)
M2: The single compartmental model (zero order)	$Cm = Yt$	Riffaldi et al. (1996);
M3 : The bi-compartimental model (exponential + linear)	$Cm = Co*(1 - exp^{-k*t}) + h*t,$	Nicolardot (1988) Houot et al. (1989)
M4 : The bi-compartimental Linear model	$Cm = CoH*t / (b.C_0H + t)$	Blet-Chraudeau et al. (1990)
M5 : The bi-compartimental model double exponential	$Cm = C1 (1 - e^{-k_1t}) + C2 (1 - e^{-k_2t})$	Delphin (1988)
M 6: Hyperbolic model	$Cm = CoH. t / (b + t)$	Juma et al. (1984)

effect of the two manures (F1 and F2) on the mineralization ( $^{14}\text{CO}_2$ ) of glyphosate and 2.4-D in the different systems compared to controls without organic matter "S1H1, S1H2, S2H1 and S2H2" over an incubation period of 150 days (Table 4, 5, 6 and 7).

In order to model and simulate the results recorded in the previous Tables (Tables 4, 5, 6 and 7) of mineralization kinetics of the two herbicides according to the XLstat software, we used nonlinear regression methods that to say the statistical analysis consisted of stand-out nonlinear correlations. For the parameter prediction equations of the six herbicide mineralization kinetics models under the effect of the two manures, non-linear adjustment was used:

1. M 1: The single-compartment model (first-order exponential)
2. M 2: The single compartmental model (zero order)
3. M 3: The bi-compartmental model (exponential + linear)

Table 4: Effects of two manures (F1 and F2) on the mineralization kinetics of  $^{14}\text{C}$ -glyphosate in the soil of Beni Ammar (S1) during 150 days of incubation.

Time (days)	S1+H1	S1+H1+F1	S1+H1+F2
0	0.100	2.000	2.500
3	3.000	9.530	16.750
7	8.750	19.220	40.890
10	13.150	34.750	64.090
14	26.120	48.170	78.080
21	28.270	59.120	82.100
28	29.190	64.920	86.790
42	30.190	72.210	89.190
60	32.130	77.120	90.230
90	70.930	79.550	91.130
120	42.37	82.21	92.21

Table 5: Effects of two manures (F1 and F2) on the mineralization kinetics of  $^{14}\text{C}$ -2,4-D in the soil of Beni Ammar (S1) during 150 days of incubation.

Time (days)	S1+H2	S1+H2+F1	S1H2F2
0	1.930	0.780	1.050
3	8.250	8.150	4.490
7	9.370	13.750	30.980
10	14.850	30.230	55.110
14	20.210	42.120	63.890
21	22.350	50.950	69.220
28	24.210	64.920	72.320
42	25.380	66.780	75.230
60	25.920	68.970	76.390
90	27.130	71.120	77.230
120	29.25	73.12	77.82

4. M 4: The bi-compartmental model (double Linear)
5. M 5: The bi-compartmental model (double exponential)
6. M 6: Hyperbolic model

The classification and selection criteria of the Models are the mean values of the indicators of the coefficient of determination  $R^2$  and the square root of the mean squares of the RMCE deviations extracted from the equations of six nonlinear regression models cited above. This model adequately describes the experimental data. The results obtained are recorded in the tables (Tables 8, 9, 10 and 11). Indeed, the chosen model has the highest mean value of  $R^2$  and contains the smallest mean value of RMCE.

### Effect of F1 and F2 on the Choice of Models (RNL) for Assessing the Biodegradation of Glyphosate and 2.4-D in the S1H1 System

For model 2 (M2) (zero order): No mathematical prediction,

Table 6: Effects of two manures (F1 and F2) on the mineralization kinetics of  $^{14}\text{C}$ -glyphosate in the soil of Maiz el Bachir (S2) during 150 days of incubation.

Time (days)	S2+H1	S2+H1+F1	S2+H1+F2
0	0.10	1.75	2.75
3	3.00	7.33	6.00
7	8.75	21.00	16.95
10	13.15	37.15	32.67
14	23.21	48.79	50.35
21	26.12	60.23	60.24
28	28.27	67.33	67.23
42	29.19	70.09	71.19
60	30.19	73.19	75.13
90	32.13	76.16	75.92
120	70.93	76.82	85.23

Table 7: Effects of two manures (F1 and F2) on the mineralization kinetics of  $^{14}\text{C}$ -2,4-D in the soil of Maiz el Bachir (S2) during 150 days of incubation.

Time (days)	S2+H2	S2+H2+F1	S2+H2+F2
0	0.03	1.25	1.35
3	1.75	4.35	6.25
7	6.38	7.39	11.79
10	11.59	16.07	27.67
14	18.76	29.93	38.21
21	20.23	44.28	51.33
28	20.82	54.33	60.23
42	22.20	60.09	64.10
60	23.55	62.12	67.92
90	25.13	63.82	70.10
120	66.28	76.01	78.13

Table 8: Effects of two manure on model simulation evaluation parameters in S1H1 system.

Treatments	Classification parameter	Selected models					
		M1	M2	M3	M4	M5	M6
S1H1	R <sup>2</sup>	0.93	/	0.92	0.44	0.93	0.90
	RMCE	199.16	/	193.98	9584.33	205.45	260.95
S1H1F1D1	R <sup>2</sup>	0.989	/	0.989	0.59	0.97	0.95
	RMCE	80.65	/	89.73	2080.67	89.013	203.20
S1H1F2D1	R <sup>2</sup>	0.95	/	0.96	0.31	0.95	0.93
	RMCE	195.61	/	444.46	2759.49	228.39	782.25

Table 9: Effects of two manure on model simulation evaluation parameters in S1H2 system.

Treatments	Classification parameter	Selected models					
		M1	M2	M3	M4	M5	M6
S1H2	R <sup>2</sup>	0.985	/	0.96	0.56	0.97	0.96
	RMCE	14.62	/	14.15	936.16	0.973	32.29
S1H2F1D1	R <sup>2</sup>	0.98	/	0.98	0.64	0.98	0.55
	RMCE	14.31	/	108.57	2141.68	116.89	123
S1H2F2D1	R <sup>2</sup>	0.973	/	0.95	0.43	0.948	0.924
	RMCE	242.195	/	493.30	5600.024	522.161	721.52

which translates into slashes in the Table (/), logical result because it is a linear function in the straight form ( $C_m = Y_t$ ). Note that the M1 model has the highest average R<sup>2</sup> values of 0.930 in the S1H1 system. However, under the influence of manure 1 (F1) and manure 2 (F2), these coefficients of determination become 0.989 and 0.95 respectively. However, the smallest mean root value of the mean square deviations noted in model M1 is 199.16, 80.65, and 195.61 for S1H1, S1H1F1D1, and S1H1F2D1 (Table 8).

About the four rest models, the choice according to the mean values of the adjustment parameters R<sup>2</sup> (0.93, 0.92, 0.90, 0.64) and RMCE ranked as follows M5, M3, M6 and M4 in S1H1. While for the two systems S1H1F1 and S1H1F2, the choice is classified in descending order M3, M5, M6 and M4 according to R<sup>2</sup> and RMCE

Finally, we conclude that the average values of R<sup>2</sup> are the highest and RMCE the lowest, the mono-compartmental

M1 model would be the model that adequately describes the experimental data.

**Effect of F1 and F2 on the Choice of Models (RNL) for Assessing the Biodegradation of Glyphosate and 2.4-D in the S1H2 System**

It was noted that M1 offers a relatively higher coefficient of determination ( $R^2 = 0.93$  in S1H2, 0.98 and 0.96 in S1H2F1 and S1H2F2 respectively) and relatively low RMCE values which leads to the conclusion that the M1 mono-compartmental (first-order exponential) is prioritized in the S1H2, S1H2F1 and S1H2F2 systems and best adjusts to our data (Table 8). By comparing the different mean values of R<sup>2</sup> and RMCE for the other models, we will conclude that these models are ranked in descending order M3, M5, M6 and M4.

However, the M2 model remains unable to simulate the mineralization kinetics of 2.4-D (H2).

Table 10: Effects of two manure on model simulation evaluation parameters in S2H1 system.

Treatments	Classification parameter	Selected models					
		M1	M2	M3	M4	M5	M6
S2H1	R <sup>2</sup>	0.93	/	0.69	0.392	0.91	0.821
	RMCE	840.306	/	44.95	3346.51	769.052	627.027
S2H1F1D1	R <sup>2</sup>	0.966	/	0.973	0.183	0.955	0.949
	RMCE	253.423	/	201.55	6372.96	229.637	361.688
S2H1F2D1	R <sup>2</sup>	0.76	/	0.66	0.23	0.55	0.54
	RMCE	1090	/	198	434	564	546

Finally, the effect of F1 and F2 on the average values of the potentially mineralizable carbon of H2 generated by the nonlinear regression of the mono-compartmental model characterized by the largest  $R^2$  (0.98 and 0.97) while the lowest RMCE (4.31 and 242.195).

### Effect of F1 and F2 on the Choice of Models (RNL) for Assessing the Biodegradation of Glyphosate and 2.4-D in the S2H1 System

The various indicators of the quality of the adjustments testify to the good-made optimizations (Table 10). The different mean of regression coefficients obtained on the dataset of M1, M5, and M6 are strong, that's to say they are the best, and the highest in M1. However,  $R^2$  for the M4 model is very low, this poor fit (M4) when it is a linear bi-compartmental, which may be due to positions related to the majority of the points being gold of the two lines.

The RMCE values decrease less in M1 than those of M5, M6, and M3, which means that the simulation by the mono-compartmental model at an ascending exponential is better than those of the others. By against, the M2 remains incapable of affecting the results, because the latter resembles a straight line (linear regression).

### Effect of F1 and F2 on the Choice of Models (RNL) for Assessing the Biodegradation of Glyphosate and 2.4-D in the S2H2 System

For soil (S2) system +2.4-D, the models have generated relatively high average  $R^2$ , either  $R^2$  is equal to 0.91, 0.90, 0.88 and 0.82 respectively for M1, M5, M6 and M3, while the M4 model is generated from very low  $R^2$  (Table 11).

Under the effect of the bovine amendment, the average values of  $R^2$  are high. Indeed,  $R^2$  is equal to 0.96, 0.95, 0.85 and 0.90 respectively for M1, M5, M6 and M3. In parallel,

under the effect of ovine, these coefficients ( $R^2$ ) are also higher, being 0.977, 0.94, 0.96 and 0.94 respectively for M1, M5, M6 and M3.

However, for the linear bi-behavioral M4 model,  $R^2$  is low under the effect of the two manures and what concerns the M2 model is still unable to simulate the effect of manure on the mineralization kinetics of the 2.4-D herbicide.

For the second parameter, we distinguish the average value of RMCE as the smallest in M1, either 185.81 (Table 11). Finally, the M1 model has a higher value of  $R^2$  and a lower value of RMCE.

For this reason, we deduce that a priori it is the M1 model which offers a better adjustment of the experimental data and which simulates well the effect of cattle manure and sheep manure on the kinetics of mineralization of glyphosate and 2.4-D. In light of the results (Tables 8, 9, 10 and 11), the mono-compartmental model with an ascending exponential is best suited to describe this biodegradation. This model is expressed by the equation:

$${}^{14}C_m = C_0 [1 - \exp(-k \cdot t)]$$

Or,

$C_m$ : Quantity of mineralized carbon in a given time  $t$ ;

$C_0$ : Quantity of easily mineralizable carbon;

$k$ : Mineralization rate constant.

## CONCLUSIONS

To simulate our experimental data, mathematical nonlinear regression methods were adjusted. We tried to test six models; indeed, we carried out with the statistical software XLSTAT 2009. These models used to predict the degradation and disappearance of herbicides mixed with manures in the agrosystem are frequently used. The two parameters of choice are the highest coefficients of determination  $R^2$  and the lowest roots of the mean of the squared deviations (RMCE) to the point of making it possible to retain the best model of the biodegradation of the two herbicides under the effect of manures in the agricultural soils of the Bounamoussa irrigable perimeter.

Table 11: Effects of two manure on model simulation evaluation parameters in S2H2 system.

Treatments	Classification parameter	Selected models					
		M1	M2	M3	M4	M5	M6
S2H2	$R^2$	0.91	/	0.82	0.30	0.90	0.88
	RMCE	185.81	/	554.05	2373.10	635.03	702.65
S2H2F1D1	$R^2$	0.96	/	0.90	0.10	0.95	0.85
	RMCE	228.13	/	224.94	6485.65	237.65	315.91
S2H2F2D1	$R^2$	0.977	/	0.94	0.203	0.94	0.96
	RMCE	183.28	/	182.56	5935.52	162.2	272.93

For the prediction equations of the parameters of the nonlinear adjustment, six models of the mineralization kinetics of the herbicides under the effect of these manures were used, which are the first-order exponential model (M1), the zero-order model (M2), The exponential + linear model (M3), the bi-compartmental Linear model (M4), the double exponential model (M5) and Hyperbolic model (M6).

The mathematical prediction model that best met these two conditions is the mono-compartmental model with a first-order ascending exponential. Although this model faithfully expresses the kinetics of the disappearance (mineralization) of glyphosate and 2,4-D in soils, the active ingredient of the herbicide needs a single degradation rate. It appears that this model is designed to reproduce reality because it gives the description, explanation, and prediction of this process.

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