Simulating the fate and transport of pesticide in unsaturated soil: a case study with glyphosate-isopropylammonium

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Abstract

A simultaneous transport of water and glyphosate was studied experimentally and numerically. A greenhouse experiment was achieved for collecting data on the water content and glyphosate concentration. A numerical model (LEACHM) was used for simulating these data. Simulated results were compared with experimental data. Experiments were conducted in one-end open PVC columns (0.056-m ID and 1-m high) using unsaturated sandy soil under the atmospheric condition of Buridah, Al-Qassium, KSA. The soil had an initial water content of 0.0802 m$^3$/m$^3$ and zero glyphosate concentration. Two soil columns lasted for 30 days and the other two for 60 days. Glyphosate solution was added at the open end of soil column then followed by adding distilled water at different times during the first 30 days. Both predicted and measured final water showed nonlinear distributions. The water moved from the hot region toward the cold region. The model overestimated slightly the water content in comparison to the observed data. The glyphosate redistributed along the soil column experimentally and numerically. The predicted glyphosate concentrations were improved by increasing the dispersivity up to 75 mm. The observed glyphosate concentration proved that the mass flow mechanism is important for migration of the glyphosate in the sandy soil. The results of this study indicated that transport models need to include the effect of temperature and temperature gradient to describe the movement of water and glyphosate. This could lead to using the LEACHM as a tool for glyphosate management strategy.

Keywords: LEACHM, glyphosate, water movement, modelling.
1 Introduction

The agricultural section around the world uses a large quantity of pesticides for protecting the field crops against insects, weeds, and plant diseases. For example, the estimated use of glyphosate in the United State of America is between 38 and 48 million pounds per year Aspelin [1]. However, glyphosate poses a serious threat to public health and many other organisms via the contamination of water resources, soil, and plant. For example, Symptoms effects of the glyphosate on the human include destruction of red blood cells, excess fluid in lungs, and pneumonia Sawada et al [2]. In laboratory tests, glyphosate increased plants' susceptibility to disease such as crown and root disease in tomatoes Brammall and Higgins [3]. In addition, glyphosate injection of lodgepole pine trees inhibited the defensive response of the trees to blue stain fungus Bergvinson and Borden [4].

Since the agricultural sector is the largest for consuming the glyphosate, we need to understand the fate and movement of this herbicide in the soil. Glyphosate’s persistence in soil varies widely, so giving a simple answer to the question "How long does glyphosate persist in soil?" is not possible. Half-lives are as low as 3 days (in Texas) and as long as 141 days (in Iowa) U.S. EPA [5]. Wauchope et al [6] reported that glyphosate is strongly adsorbed to most soils, even those with lower organic and clay content, with field half-lives range from 1 to 174 days. However, glyphosate can move into surface water when the soil particles to which it is bound are washed into streams or rivers U.S. EPA [7]. The soil reaction pH is a major factor affecting the adsorption characteristic of glyphosate in a soil. Eberbach [8] found that glyphosate appeared to be more strongly held in the acid Rutherglen soil than in the alkaline soils. The author attributed this difference to the lower pH and higher iron content of the acid soils. Even though it is highly soluble in water, field and laboratory studies show it does not leach appreciably, and has low potential for runoff (except as adsorbed to colloidal matter) Wauchope et al [6]. Microbes are primarily responsible for the breakdown of the product, and volatilization or photodegradation losses will be negligible Weed Science Society of America, Herbicide Handbook [9].

Glyphosate can move by several mechanisms. Therefore, understanding the mechanisms of movement of the glyphosate can lead to better management. A model describing solute (inorganic and organic) fate and movement, water, and heat movement in porous media was developed by Hutson and Wagenet [10]. The model is called LEACHM. The model includes matric water pressure head, gravity pressure head, and solute concentration gradients as driving forces for water and solute transfer. The organic compounds can be associated with liquid and solid phases of the porous media. The liquid phase is mobile, but the solid phase is immobile. The transfer of organic compounds across the gas/liquid, liquid/solid, and gas/solid interfaces is included using equilibrium assumptions at the interfaces. Under the condition of the present study, transfer across liquid/solid is considered only because the glyphosate is not volatile. The degradation of glyphosate is described by a first-order decay rate. The model can
be used to predict spatial and temporal variations of water content, and the total concentration of solute within a porous medium.

According to the literature cited above, glyphosate movement in soil has rarely been studied experimentally under nonisothermal conditions. The research project encompassed greenhouse and theoretical analyses. Therefore, the first objective of this study is to perform nonisothermal experiments under greenhouse to observe water, and glyphosate transfer in unsaturated soil. The second objective is to test model of LEACHM in partially saturated and unsaturated soil conditions against the greenhouse experimental data.

2 Materials and methods

2.1 Experimental

Soil materials were sampled from a surface layer (0.0-0.3 m depth) from the Agriculture and Veterinary Collage farm, King Saud University, Al-Qassim. The soil materials were obtained by excavation, then air-dried and ground to pass a 2-mm sieve. The soil was composed of sand (96.3 % sand, 1.9 % clay, and 1.8 % silt) materials. The resulting soil materials were wetted with distilled water to obtain an initial volumetric water content of 0.0802 m³/m³. The moistened soil was covered and stored at 20 °C for 4 days. PVC cylinders (0.056-m ID and 1-m high) were closed at the bottom ends using epoxy-sealed PVC lids then packed with the moistened soil each 0.05-m increment. Four soil columns were packed at bulk densities of 1514 kg m⁻³. The soil columns were buried vertically within a bare soil field with exposing the upper end to the natural atmosphere of Al-Qassim region. Water and glyphosate solution were poured at the open of soil column at different time as shown in Table 1.

Table 1: Amount and time of water or glyphosate solution application (m/column).

<table>
<thead>
<tr>
<th>Amount</th>
<th>0.0203</th>
<th>0.0406*</th>
<th>0.0203</th>
<th>0.0203</th>
<th>0.0203</th>
</tr>
</thead>
</table>

The soil temperatures at both ends of soil column were recorded. Two soil columns were removed and sectioned after 30 days of the experiment initiation and the other two columns were sectioned 30 days later. The soil columns were sectioned into a 0.05-m increment. The soil of each increment was divided into two parts: a part for water content and a part for glyphosate determinations. The soil water contents were determined gravimetrically in the increments. The glyphosate was measured in second part of the soil sample. For glyphosate determination, soil was immediately kept at -20°C until analysis. The organic phosphate in glyphosate was oxidized with hydrogen peroxide to the orthophosphate, which measured calorimetrically as the phosphomolybdate heteropoly blue complex at 830 nm Glass [11]. The absorbance was recorded...
after at 830 nm using UV/Visible Spectrophotometer. Calibration curve was performed over 0-100µg/ml\(^1\) (Figure 1). The relation between the absorbance density and the concentration of glyphosate was linear with determination coefficient of 0.99. The recovery of the analysis was 96-105 % of applied amount for glyphosate. These values are in range reported by Autio et al [12].

Figure 1: Calibration curve for the concentration of glyphosate vs. the optical density.

2.2 Theoretical analysis

LEACHM model devolved by Hutson and Wagenet [10] was used to describe water and glyphosate transfer in a soil. The model has several options of describing water flow. The mechanistic Richards equation option was used in the present study. Heat moves by conduction mechanism. The solute flow based upon the convection-dispersion equation (CDE) was used for describing the
glyphosate flow. The finite differences scheme for the partial differential equations of heat, water, and solute transfer was used in the model. A brief outline of LEACHM is presented below for completeness, with additional details found elsewhere Hutson and Wagenet [10].

2.2.1 Water flow
LEACHM uses a finite-difference form of Richard’s equation as a means of predicting water contents, fluxes and potentials. Richard’s equation, the soil water flow equation for transient vertical flow derived from Darcy’s law and the continuity equation, is:

$$\frac{\partial \theta}{\partial t} = \frac{\partial \psi}{\partial t} C_w = - \frac{\partial q}{\partial z} - U(z,t)$$

(1)

where \( \theta \) is water content (\( L^3 \) \( L^{-3} \)), \( q \) is water flux density (\( L/T \), defined as \( q = -K(\Psi)(\delta H/\delta z) \)), \( H \) is hydraulic head (\( L \), defined as \( H = \Psi - z \), \( \Psi \) is soil water matric potential and \( z \) is depth), \( K \) is hydraulic conductivity (\( L/T \)), \( t \) is time (\( T \)), \( C_w \) is differential water capacity (\( L^{-1} \)), and \( U \) is a sink term representing water lost per unit time by transpiration (\( T^{-1} \)). The later term is neglected under the present study because there is no plant. The water transfer properties (\( \Psi - \theta \), and hydraulic conductivity, \( K \)) were described in detail Campbell [13].

2.2.2 Pesticide flow and transformation
The movement of miscible pesticide (i.e., glyphosate) through a soil is assumed in LEACHM to be accomplished by chemical diffusion in the liquid phase in response to an aqueous concentration gradient and convection of the pesticide as the result of movement of water flow in which the pesticide is dissolved. That is:

$$J_t = J_d + J_c$$

(2)

Where \( J_t \) is total pesticide flux (\( M/L^2 T \)), \( J_d \) and \( J_c \) are diffusion and convection fluxes in the liquid phase, respectively.

The diffusion flux in a soil solution can be obtained from:

$$J_d = -D_p(\theta) \frac{\partial C_l}{\partial z}$$

(3)

where \( D_p(\theta) = D_o \varepsilon \exp(b*\theta) \), is the diffusion coefficient of pesticide in porous media and \( \varepsilon \) and \( b \) are empirical constants Olsen and Kemper [14]. Values of \( a \) ranged from 0.05 to 0.01 and \( b = 10 \). \( C_l \) is the concentration in the liquid phase (\( M/L^2 \)). \( D_o \) is the diffusion coefficient of a pesticide in a free-water system (\( L^2/T \)). The diffusion coefficient was described in details Nassar and Horton [15] and Lyman et al [16].

The convective flux of a pesticide is usually represented as:
\[ J_c = -\theta D_m(q) \frac{\partial C_l}{\partial z} + qC_l \]  

(4)

where \( D_m(q) = \lambda |v| \), is the mechanical dispersion coefficient that describes mixing between large and small pores, \( v = q/\theta \), is the average pore velocity and \( \lambda \) is the dispersivity (L) and its value ranged between 0.5 \( \Delta z \) to 2 \( \Delta z \), and \( \Delta z \) is node spacing (L).

The total steady state solute flux in the liquid phase in a porous media, \( J_t \) can be described as:

\[ J_t = -\theta D_m(q) \left( \frac{\partial C_l}{\partial z} \right) - D_p(\theta) \frac{\partial C_l}{\partial z} + qC_l \]  

(5)

The nonsteady-state equation for the pesticide transport can be written as:

\[ \frac{\partial C_t}{\partial t} = -\frac{\partial J_t}{\partial z} \pm \Phi \]  

(6)

Where \( C_t = \theta C_l + \rho_b C_s \), is the total pesticide concentration in liquid and sorbed phases (M/L³), \( \rho_b \) is the bulk density of soil (M/L³), \( C_s = K_d C_l \), is the concentration of pesticide in the sorbed phase (M/M), \( K_d \) is a partition coefficient (L³/M), and \( \Phi \) is sources or sinks of pesticide (such as microbiological degradation) (M/L³ T).

The partition coefficient, \( K_d \) can be estimated as

\[ K_d = K_{oc} f_{oc} \]  

(7)

Where \( K_{oc} \) is the organic carbon partition coefficient (L³/M) and \( f_{oc} \) is the organic carbon fraction.

The \( K_{oc} \) can be estimated as [16]:

\[ \log(K_{oc}) = -0.557 \log(S) + 4.277 \]  

(8)

Where \( S \) is the solubility of pesticide in water (µmol/l).

Degradation of total pesticide is assumed to obey first-order kinetics, so that

\[ \Phi = -k(\theta + \rho K_d)C_l \]  

(9)

where \( k \) is a first − order rate coefficient (T⁻¹).

**2.2.3 Initial and boundary condition**

The initial conditions associated with Eqs. (1), (2), (3), and (4) are given by

\[ \theta(z,0) = \theta_i, \quad C_t(z,0) = 0.0 \]  

(10)

The upper boundary conditions for water and solute are given in terms of net mass fluxes by
\[ q(0, t) = \zeta E \]  \hspace{1cm} \text{(under the evaporation conduction)} \hspace{1cm} (11)

\[ J_i(0, t) = 0.0 \]  \hspace{1cm} \text{(for a non-volatile compound)} \hspace{1cm} (12)

or

\[ q(0, t) = \inf \]  \hspace{1cm} \text{(under ponding condition)}

\[ J_i(0, t) = q(0, t)C_i \]  \hspace{1cm} (13)

where \( \zeta \) is the evaporation pan coefficient, \( E \) is the potential evaporation rate (LT\(^{-1}\)), \( \inf \) (LT\(^{-1}\)) is the infiltration rate and \( C_i \) is concentration of a compound in the applied water (M/L\(^3\)). In the present study, \( C_i \) for glyphosate was 48x10\(^4\) ppm.

The lower boundary condition for water and glyphosate are:

\[ q(l, t) = 0.0 \]  \hspace{1cm} (14)
\[ j_i(l, t) = 0.0 \]  \hspace{1cm} (15)

### 2.3 Model and results evaluation

Model ability to predict a variable should be evaluated. Visual comparison of simulated and observed data provides a quick and often comprehensive mean of assessing the accuracy of model prediction. However, quantitative evaluation of the model is recommended. In the present work, the mean error (ME) and root mean square error (RMSE) are used as criteria for evaluating the model Milly [17]. The ME values can be estimated as:

\[ ME = \frac{1}{N} \sum_{i=1}^{N} (V_p - V_m) \]  \hspace{1cm} (16)

and RMSE can be estimated as:

\[ RMSE = \left( \frac{1}{N} \sum_{i=1}^{N} (V_p - V_m)^2 \right)^{1/2} \]  \hspace{1cm} (17)

\( V_m \) is the observed value of a variable (\( \theta \), or \( C_i \)), \( V_p \) is the predicted value of a variable, and \( N \) is number of observations for the variable.

### 3 Results and discussion

#### 3.1 Soil parameters and characterization

The retention curve data (\( \Psi \) vs. \( \theta \)) and the saturated hydraulic conductivity were measured for the sandy soil used in the present study. The relation of \( \Psi \) vs. \( \theta \) was first described using Van Genuchten’s model [18] then the resulted values were fitted to the Campbell's function. The coefficients of the function, \( \Psi_e \) and \( b \), are shown in Table 2. Their values are in the expected range for the sandy soil Campbell [13]. The resulted and calculated retention curves are shown in Fig. 2. with a determination coefficient, \( R^2 \), = 0.98. The logarithmic calculated values of
the retention curve using the Campbell’s model showed linearity. The saturated hydraulic conductivity, \(K_s\), of the soil was measured in a laboratory soil column using a constant head method Klute and Dirksen[19]. A value of \(2.25 \times 10^{-5}\) m/s for \(K_s\) was obtained (Table 2). The unsaturated hydraulic conductivity, \(K(\Psi)\), was estimated from knowledge of \(K_s\) and the retention curve data Campbell [13]. The unsaturated hydraulic conductivity, \(K\), as a function of soil matric potential for the sandy soil is shown in Fig. 3. The values of \(K\) increased abruptly at \(\Psi\) of \(-500\) kPa with further increasing in \(\Psi\). At \(\Psi\) less \(-500\) kPa the \(K\) values were small. Similar calculated values of \(K\) are reported in Scanlon and Milly [20].

Figure 2: The soil moisture retention curve for the sandy soil.
Table 2: Input characterization data for LEACHM.

<table>
<thead>
<tr>
<th>Parameters</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>ρ (bulk density) (Mg m⁻³)</td>
<td>1.514</td>
</tr>
<tr>
<td>θᵢ (initial water content) (m³ m⁻³)</td>
<td>0.0802</td>
</tr>
<tr>
<td>θₛ (saturated water content) (m³ m⁻³)</td>
<td>0.4286</td>
</tr>
<tr>
<td>Kₛ (m⁻¹)</td>
<td>2.25x10⁻³</td>
</tr>
<tr>
<td>Clay (%)</td>
<td>1.9</td>
</tr>
<tr>
<td>Sand (%)</td>
<td>96.3</td>
</tr>
<tr>
<td>Silt (%)</td>
<td>1.8</td>
</tr>
<tr>
<td>Kₒc (m³ kg⁻¹)</td>
<td>3.68x10⁻¹²</td>
</tr>
<tr>
<td>Dₒ (m² s⁻¹)</td>
<td>6.478x10⁻¹⁰</td>
</tr>
<tr>
<td>Fₛ (%)</td>
<td>0.05</td>
</tr>
<tr>
<td>k (s⁻¹)</td>
<td>1.16x10⁻⁶</td>
</tr>
<tr>
<td>Ψₑ (kPa)</td>
<td>-0.38</td>
</tr>
<tr>
<td>B</td>
<td>1.81</td>
</tr>
<tr>
<td>S (kg/m³)</td>
<td>1050</td>
</tr>
<tr>
<td>Saturated vapor density (kg/m³)</td>
<td>2.06x10⁻¹⁰</td>
</tr>
</tbody>
</table>

3.2 Predicted and measured soil status

The predicted and measured soil water content distributions in nonisothermal columns are shown in Fig. 4. for 30 and 60 day periods. The predicted water content distributions matched the observed values only in trend. For 30 days-period, there are two distinguished zones for the distribution (observed and predicted data). The first zone extended to 0.6 m –depth and the second zone was below 0.6m –depth. The water content in the first zone was less 0.13 m³/m³ (the mean soil water content within soil column) while the second zone possessed water content greater than 0.13 m³/m³, it is expected that water movement in the upper zone mainly in vapor phase Nassar et al [21]. Increasing the water content in the second zone could be due to matric potential and temperature gradient as a driving forces for water transfer. Similar trend was obtained for data of 60-day period. The first zone extended to a 0.75-m depth which has water content less 0.115 m³/m³ (the mean soil water content within soil column). The mean soil water contents calculated from the measured data and predicted data were 0.132 and 0.151 m³/m³, respectively, for 30 day period. The predicted soil water mean is greater than the measured mean because the LEACHM model neglected the temperature gradient effect on water movement. Several researchers found that temperature gradient has great effect on water flow Globus [22] and Nassar et al [21]. The mean soil water content for the measured and predicted water contents were 0.115 and 0.141 m³/m³, respectively. In general, the data of 60 day period are in a similar trend and support the aforementioned data. The predicted and measured water contents in the upper zone (0.6 m) for the data of 60 day period were lower than that for the data of 30 day period. The difference in the water content between the two periods is due to water evaporation losses. The water contents changed abruptly in that zone for the data of 30 days period while changed gradually for 60 days period.
Figure 3: The unsaturated hydraulic conductivity for the sandy soil.

Figure 5 shows the distribution of predicted soil water matric potential for 30 and 60 days periods. The predicted matric potential for 30 days period was greater than its values for the 60 days period. These results support the water content distribution shown in Figure 4. The matric potential increases as the depth increases for both periods. The matric potential changes abruptly in the upper zone and these changes were greater for the periods of 60 d. Therefore, the water and glyphosate movement occurs mainly in this zone.
3.3 Predicted and measured glyphosate concentration

Measured and predicted Glyphosate concentration are compared in Fig. 6 for time 60 d. Despite good agreement between the measured and simulated water distributions, there is substantial disagreement between measured and simulated glyphosate. This discrepancy is apparent great in the simulated data after 30 d (data are not shown). One reason for the disagreement could due to the assumption that degradation of total pesticide obeys first-order kinetics. Similar results were reported by Wagenet et al [23] for simulation of DBCP (1,2-dibromo-3-chloropropane). The variation between the simulated and measured glyphosate was reduced by increasing the value of solute dispersivity. Three values for dispersivity
were selected and ranged from 25 –75 mm. Improving the simulation as a result from increasing the dispersivity can lead to the dominate of glyphosate movement by mass flow over the diffusion flow. In general, the measured value of glyphosate showed little difference within the soil column. This results support the aforementioned conclusion for the dominant of mass flow mechanism for solute transfer. Veiga et al [24] reported similar results and they attributed this results to the high solubility of glyphosate together with the high conductivity of soil. In addition, the distribution coefficient (K_d) of glyphosate is small (0.343 x 10^{-3} m^3/kg). Since the K_d in the present study is low, most of the glyphosate exist in the mobile phase which enhances its movement with mass flow of water. The total measured and predicted glyphosate concentration means were 40.91 and 49.79 ppm, respectively. In comparison with an initial glyphosate of 283.56 ppm, a great amount of glyphosate degraded after 60 days. The higher dose of glyphosate to be added, the faster degradation by microbial occurs.

![Predicted matric potential after 60 days of the experiment initiation.](image)

Figure 5: Predicted matric potential after 60 days of the experiment intiation.
In a phosphorous-poor soil, e.g. sandy soil, glyphosate can be source for the microbial activity. The total glyphosate concentration was negligible after one month of its application in a forest soil in Spain Veiga et al [24]. In addition, the glyphosate and its main metabolite exhibited high vertical mobility in the treated soil. The favorable fate of a pesticide is a combination of fast degradation and strong adsorption to soil. Besides sorption, other mechanism, e.g. preferential flow through macropore in soil, contributes to leaching of pesticides. In three experimental studies, Kladivko et al [25]; Traub-Eberhard et al [26]; and Brown et al [27], the rank-order in pesticides leaching corresponded to the rank-order of the sorption coefficient ($K_{oc}$). The most sensitive parameters of pesticide models are related the sorption and degradation of the compounds to be studied Autio et al [12]. Those authors reported that glyphosate could be classified as immobile if the $K_{foc}$, calculated from Freundlich Eq., exceeding 5 m$^3$/kg. Paradoxically the amount of labile glyphosate ranged from 24.1 to 34.5 % while the non-labile glyphosate ranged from 67.2 to 74.9 % in a soil with high organic fraction Eberbach [8].

Figure 6: Observed and predicted glyphosate distribution after 60 days of experiment intiation. The predicted value at 25,50 and 75mm dispersivity.
3.4 Model evaluation

Table 3 shows the calculated mean error (ME) and root mean square error (RMSE) for the water content (Fig. 4) and glyphosate (Fig. 6). For glyphosate results, the predicted values were obtained for with different dispersivity values. The ME values were 0.0186 and 0.0256 m³/m³ for 30 and 60 days data, respectively. The ratio between ME and the measured mean of soil water content were 0.14 and 0.22 for 30 and 60 days, respectively. It can be concluded that the performance in wet soil (i.e., 30 day data) is better than in dry soil (i.e., 60 day data). Similar trends for the RMSE were obtained for the predicted soil water contents. The ME values were 10.92, 9.17, and 8.88 ppm for predicted glyphosate concentration with dispersivity of 25, 50, and 75 mm, respectively. These values represented 26.7, 22.4, and 21.7 % from the corresponding measured mean concentration of glyphosate, respectively. Therefore, increasing the dispersivity improved the predicted values. The corresponding RMSE values were 30.21, 17.36 and 11.43 ppm for the predicted glyphosate concentration (Fig. 6). It is obviously, increasing the dispersivity reduced discrepancy between the observed and predicted glyphosate concentration. It can be concluded that the model described the mechanisms of water and glyphosate flows appropriately under the conditions of the present study.

<table>
<thead>
<tr>
<th>ME or RMSE</th>
<th>Fig. 4a</th>
<th>Fig. 4b</th>
<th>Fig 6 ($\lambda$ in mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>ME</td>
<td>0.019</td>
<td>0.025</td>
<td>25</td>
</tr>
<tr>
<td>RMSE</td>
<td>0.012</td>
<td>0.017</td>
<td>10.92</td>
</tr>
</tbody>
</table>

4 Summary and conclusion

A greenhouse experiment was achieved for monitoring water and glyphosate transfer by using one-end-open soil columns. The soil columns were buried vertically in a bare soil field in Al-Qassium region, KSA. Glyphosate solution was added to unsaturated soil four days after the experiment initiation. The LEACHM model of heat and mass transfer was used to predict water content and glyphosate within the soil columns. The predicted and observed water content distribution was in agreement. The predicted mean of soil water content was greater than the observed one. The over predicted mean water content might be due to neglecting the soil temperature gradient on water transfer. There was a discrepancy between the predicted and observed Glyphosate after 60 days of the experiment initiation within the soil columns. Increasing the dispersivity values to 75 mm reduced the discrepancy. The predicted values peaked at 0.3-m depth while the observed did not. The application of this model may be very useful for the development of remediation strategies to clean soil from glyphosate compounds when nonisothermal conditions exist and the degradation, and sorption coefficient are...
measured for the studied site. It should also be useful for monitoring glyphosate redistribution at contaminated sites.

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**References**


