

Gas-Diffusivity based characterization of aggregated agricultural soils

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Abstract

Grazed pastures and cultivated fields are significant sources of greenhouse gas (GHG) emissions, in particular N₂O emissions derived from fertilizer deposition and animal excreta. Net surface emissions rely on subsurface gas transfer controlled mainly by diffusion, expressed as the soil-gas diffusivity (D_p/D_o). The value of D_p/D_o is a function of soil air-filled porosity (ϵ) and gaseous phase tortuosity (τ), both of which vary with soil physical properties including soil texture and structure. Agricultural soils are often structurally aggregated and characterized by two distinct regions (inter- and intra-aggregated pores), however, such soils are subjected to frequent compaction and tillage resulting in alteration to structural arrangement. In this study, a comparative analysis between the Currie (1960) and Taylor (1949) methods was performed to provide a computational insight into selecting an appropriate method for calculating D_p/D_o in agricultural soils. Currie's (1960) method was chosen for further analysis of the soils in this study. Results show that the D_p/D_o in aggregated soil cannot be expressed using a simple linear, power law or combined linear and power law functions due to the presence of two-region characteristics. A new "Two-Region model" was developed to parameterize the D_p/D_o of aggregated soils, and tested against repacked samples from two Sri Lankan agricultural soils. This Two-Region model clearly distinguished tortuosity effects on gas movement with respect to density and textural variations within and between aggregates and outperformed previous models. The fitting parameters (α_1 , α_2 , β_1 and β_2) varied correspondingly with soil density, and the weighting factor (w) clearly distinguished the boundary between the two regions (inter- and intra-aggregates) of structured soils. The model developed will be of interest to those seeking to model the diffusion of GHG emissions and gas exchange between the atmosphere and soils.

1 | INTRODUCTION

Agricultural ecosystems, including croplands and grazed pasture, are major sources of anthropogenic greenhouse

gases (GHG) generating 16% of the total global greenhouse gas emissions footprint (Andersen & Petersen, 2009). Consequently, emphasis has been given to cropland and grazed pasture management in global efforts to mitigate excessive

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agricultural GHG emissions (IPCC, 2014). Soils within these agricultural systems are generally considered to be well-structured aggregated soils, typically characterized by two distinct pore regions: pore space between aggregates referred to as ‘inter-aggregate pores’ and pore space within aggregates referred to as ‘intra-aggregate pores’, resulting in a bimodal soil pore structure. However, different land management practices, such as tillage and soil compaction, may lead to frequent re-structuring of pores, thus affecting the total porosity and pore structure. Tillage facilitates soil aeration, root growth and nutrient utilization while the requirement of oxygen (O_2) during the different stages of plant growth varies and is crop type dependent (Grable & Siemer, 1968; Stepniewski, 1980). In contrast with tillage, compaction often results in decreased porosity and deteriorated aeration, although soil-water retention properties may improve (Stepniewski, 1980). Compaction in agricultural soils may occur as a result of, for example, animal treading of pasture soils at high stocking rates or excessive use of agricultural implements (Schjønning et al., 2009; Stepniewski, 1980). Notably, both tillage and compaction make a significant impact on soil functional structure thereby affecting the aggregated status of the soil.

In addition, agricultural soils vary largely with respect to the soil texture and/or soil type, land use, and organic matter content, the latter also controls the aggregated nature of the agricultural soils. Furthermore, higher carbon inputs by livestock manure, soil-moisture dynamics due to wetting and drying during rainfall and irrigation, root exudation in the plant rhizosphere, soil fauna (e.g., earthworms) and microbial functions may all potentially help transform agricultural soils to aggregated bimodal structures (Ghezzehei, 2012; Six et al., 2004). Since a complex combination of many soil physical variables play diverse roles in aggregation, and the resulting bimodal soil structure development, there remains a need to accurately characterize agricultural soils with respect to gas migration in order to better understand the role of soil aggregation on soil gas transport characteristics.

Greenhouse gas migration in the subsurface and resulting emission to the atmosphere across the soil-atmosphere continuum occurs primarily by diffusion. Similarly, the diffusion-controlled exchange of gases that occurs during soil aeration ensures efficient transport of O_2 from the atmosphere to the O_2 -depleted plant root zone and the rapid exit of carbon dioxide (CO_2) from root zone to the atmosphere. Soil-gas diffusivity, D_p/D_o where D_p (m^3 soil air m^{-1} soil s^{-1}) and D_o (m^2 air s^{-1}) are the soil-gas diffusion coefficients for a given gas diffusing within a porous medium and free air, respectively, is the most important parameter describing diffusion-controlled gas migration in soil. The value of D_p/D_o is a function of the air-filled porosity, and the tortuosity of the functional soil gaseous phase. As such it is strongly dependent on soil physical properties such as soil texture/type, structure, total porosity, moisture content and

Core Ideas

- A ‘two-region model’ was parameterized for gas-diffusivity of aggregated soils.
- The model distinguished tortuosity effects of density and texture on diffusivity.
- The model performed better than previously recognized models.
- The model accounted well for inter-aggregate and intra-aggregate effects.
- The developed model will assist those seeking to understand soil gas exchange.

organic matter content (Chamindu Deepagoda et al., 2011a; Resurreccion et al., 2007). Recent studies have shown strong correlations between measured D_p/D_o and N_2O fluxes in agricultural soils (e.g. Balaine et al., 2013; Owens et al., 2017), thus recognizing the potential of D_p/D_o as a key predictor for soil conditions that are conducive for N_2O fluxes. Recent studies have also indicated the presence of a critical diffusivity window ($D_p/D_o \sim 0.005$ – 0.01), in both intact and repacked pasture soils, which yields a peak in N_2O emissions regardless of the soil texture, structure, and soil moisture status (Chamindu Deepagoda et al., 2019).

Calculation of D_p/D_o essentially requires solving the classical Fick’s laws of diffusion under specified boundary conditions. Both Taylor’s method (Taylor, 1949) and Currie’s method (Currie, 1960) have been commonly used to compute D_p/D_o under non-steady state measurement conditions. While the method of Taylor (Taylor, 1949) only invokes Fick’s first law of diffusion, the method of Currie (1960) uses Fick’s second law, which accounts for the production, consumption and storage of a gas in porous media (Fujikawa & Miyazaki, 2005; Rolston & Moldrup, 2002). Consequently, in highly porous media such as aggregated soils, which may potentially store larger amounts of gas compared to unimodal soils, the results of the two methods may deviate and result in erroneous results for emission estimates. Therefore, a comparative study with computations from both methods may provide a useful insight for selecting an appropriate method for further examining D_p/D_o in agricultural soils.

Measurement of D_p/D_o requires specific apparatus and controlled boundary conditions thus, it is common to use predictive models to estimate D_p/D_o from easy-to-measure soil physical parameters, for example air-filled porosity and total porosity. While predictive models are currently available to predict D_p/D_o from soil air-filled and total porosities in non-aggregated soils (e.g., Millington & Quirk, 1961; Moldrup et al., 2000, 2013), the use of these models for aggregated soils may result in biased results due to the presence of two distinct pore regions in aggregated soils. Two-region D_p/D_o

TABLE 1 Measured soil physical parameters belonging to pasture soil at the University of Peradeniya, Sri Lanka (PD-P) and arable soil at Meewathura, University of Peradeniya, Sri Lanka (PD-C), and two agricultural soils from Tokyo, Japan: a Nishi-Tokyo cultivated soil (NT-C) and a Nishi-Tokyo pasture soil (NT-P)

Sampling site	Sampling depth	Soil class	Moisture content	Porosity	Particle density	In situ density	Dry density	Organic content
			%	$\text{cm}^3\text{cm}^{-3}$	g cm^{-3}	g cm^{-3}	%	
PD-P	0–10 cm	Sandy loam	22.07	0.463	2.59	1.64	1.33	9.58
PD-C	0–5 cm	Sandy loam	11.41	0.471	2.68	1.59	1.42	4.60
	5–10 cm	Sandy loam	12.50	0.518	2.63	1.44	1.26	4.80
	10–15 cm	Sandy loam	14.11	0.542	2.64	1.41	1.23	4.50
NT-P	0–10 cm	Silt loam	NA ^a	0.76	NA	NA	NA	11.0
NT-C	0–15 cm	Silt loam	NA	0.74–0.76	NA	NA	NA	NA

^aNA, not available

models, consisting of empirical (e.g., Resurreccion et al., 2008b, 2010), as well as theoretical (e.g., Ghnabarian et al., 2014; Hunt et al., 2014) models have been proposed in the literature, but these have only been applied to a limited extent in attempts to adequately characterize D_p/D_o in aggregated soils. Thus, more modelling efforts are required to accurately characterize two-region D_p/D_o behavior in agricultural soils, with minimum soil physical parameter inputs.

The main objective of this study was to perform a gas diffusivity-based characterization of repacked aggregated soils sampled from both a grazed pasture and a cultivated land in Sri Lanka. Specifically, we investigated the effect of soil structure/aggregation on diffusion-controlled gas migration in the two selected sites which constituted a distinct soil structural contrast that resulted from compaction due to animal treading (pasture) and frequent tillage (cultivated land). In this study, we designated the soils with a distinct two-region (bimodal) pore structure as “aggregated soils” to distinguish them from the non-aggregated (unimodal) soil. We further tested the two widely accepted methods for computing soil-gas diffusivity in unimodal soils, Taylor (1949) and Currie (1960), and compared them against aggregated (bimodal) soils. Further, a two-region model is proposed and parameterized with measured D_p/D_o data in differently structured aggregated soils.

2 | MATERIALS AND METHODS

2.1 | Sampling sites, soils, and data

Disturbed soil samples were collected in bulk from a pasture soil from the 0–10 cm depth of a grazed pasture site, where cattle rearing is the primary livestock activity, at the University of Peradeniya, Sri Lanka (hereafter referred to as PD-P). The soil was subjected to daily livestock treading, although agricultural machinery traffic was rare.

Disturbed soil samples were taken at three depths: 0–5, 5–10, and 10–15 cm from an arable site at Mee-

wathura, University of Peradeniya, Sri Lanka (hereafter referred to as PD-C). Previously, banana was cropped twice a year for 3 years. Prior to sampling, the site had been manually tilled and prepared for planting. Soil was specifically sampled after tillage for low-dense soils in order to capture a density variation from compacted pasture soils.

Intact soil samples of 100 cm^3 were also taken at both sites by carefully driving annular cores in to the soil to determine the in situ bulk density at respective layers. To prepare repacked cores, sampled soil clusters were manually broken to retain their micro-aggregate structures under in situ moisture condition, air dried and sieved to the desired particle size fraction ($<2\text{ mm}$). The aggregates were uniaxially packed in 100-cm^3 annular cores to the dry density values measured in situ, with an additional soil bulk density of 1.0 g cm^{-3} which resembled the lowest density observed across the pasture soil. Care was taken not to disturb the micro-aggregated nature of soils while packing. In total, 150 measurements (5 densities \times 10 moisture levels \times 3 replicates) from both sites were considered.

In order to compare the two D_p/D_o computational methods (Taylor, 1949; Currie, 1960), and to validate the introduced two-region model, we further considered data from Tokyo, Japan representing two agricultural soils: a Nishi-Tokyo cultivated soil (referred to as NT-C) sampled at 0–15 cm, and a Nishi-Tokyo pasture soil (referred to as NT-P) sampled at 0–10 cm (data from Chamindu Deepagoda et al., 2011b). Both sampling sites belonged to the Field Production Science Centre at the University of Tokyo, Japan. The soil is an Andisol of volcanic origin, and consists of significantly different textural and structural characteristics compared to the two Sri Lankan soils. Due to its allophane dominated hollow spherical morphology, the Andisol has a high total porosity and low bulk density which enables free movement of water and air through the structure. The basic soil physical properties of the two Sri Lankan soils (this study) and the two Japanese soils (from Chamindu Deepagoda et al., 2011b) are shown in Table 1.

2.2 | Measurement methods

2.2.1 | Soil-gas diffusivity (D_p/D_o)

Repacked samples were saturated for 72 hours before being subjected to stepwise air drying to obtain the intended moisture contents of moisture reduction by 5 g in each drying step for D_p/D_o measurements. The samples were then kept sealed for 24 hours to allow hydraulic equilibrium to be attained prior to each D_p/D_o measurement (Currie, 1984). To measure D_p/D_o , the one-chamber method introduced by Taylor (1949) and developed further by Schjønning (1985) was adopted. Initially the air-tight diffusion chamber was flushed with 99.99% N_2 gas to remove all O_2 inside the chamber. The sample, mounted on the chamber, was then opened to the atmosphere by allowing the atmospheric O_2 to diffuse through the sample to the chamber. The increasing O_2 concentration within the chamber was continuously monitored with an O_2 sensor (KE-25, Figaro Inc.) attached to the chamber wall. The time taken for completion of diffusion (a few minutes up to 3 hours, depending on the moisture content) is fast compared to O_2 consumption within the sample, thus changes in O_2 concentrations are considered to be only dependent on transport (Schjønning et al., 1999). To calculate the value of D_p/D_o , we used the methods of both Taylor (1949) and Currie (1960) as detailed below.

For determination of D_p/D_o in Nishi-Tokyo soils, sieved size fractions (<2 mm) were repacked in 100-cm³ annular cores at the designated total porosity values as stated in Table 1. Saturated samples were drained sequentially inside sand boxes to achieve the desired matric potentials of -1.0, -1.5, -1.8, -2.0, -2.5, -3.0, -4.1, -6.0, and -6.9 using hanging water column (for $\psi > -3$ cm H_2O or pF 1.5) and pressure plate apparatus (for $\psi < -3$ cm H_2O) for gas diffusivity measurements, which were conducted using the same one-chamber method as used for Sri Lankan soils described above. Calculation of D_p/D_o was performed following both Currie (1960) and Taylor (1949) using methods as outlined by Chamindu Deepagoda and Elberling (2015).

2.2.2 | Particle size distribution

Particle size distribution of sampled soils were measured following the wet sieving method and hydrometer test (BS 1377 Part 2; British Standards Institution, 1990). To characterize and quantify the measured particle size distribution, we invoked the extended Rosin-Rammler (1933) particle size distribution function introduced by Chamindu Deepagoda et al. (2018). The function parameterizes the percentage of particles passing (by weight), P (%), expressed as a function of

grain size, x (mm), using two fitting coefficients as follows:

$$P(x) = 100 \left\{ w \left[1 - e^{-\left(\frac{x}{\mu_f}\right)^{\sigma_f}} \right] + (1-w) \left[1 - e^{-\left(\frac{x}{\mu_c}\right)^{\sigma_c}} \right] \right\} \quad (1)$$

where μ_f and μ_c (μm) are characteristic sizes (i.e., 63rd percentile values) representing the fine and coarse size distributions, and σ_f and σ_c (dimensionless) are model fitting coefficients representing the spread of the fine and coarse grain size distribution, respectively. The weighted mass fraction of coarse particles, w (dimensionless), is also a fitting parameter together with the above parameters.

The mean particle size (the size corresponding to 50% passing), D_{50} (μm), can be derived from the model fitting parameters as follows:

$$D_{50} = \mu_i \cdot (\ln 2)^{1/\sigma_i} \quad (2)$$

where μ_i and σ_i represent corresponding parameters for fine and coarse fractions.

2.3 | Soil-gas diffusivity modeling

2.3.1 | Non-aggregated soils

Starting from the pioneering work of Buckingham (1904) a wide range of D_p/D_o models have been proposed for non-aggregated (one-region) soils which use air-filled porosity as the only model parameter (Marshall, 1959; Millington, 1959; Penman, 1940). A series of two-parameter models, which used both total and air-filled porosities, were later proposed in order to better account for soil type and additional soil moisture effects (e.g., Millington & Quirk, 1961; Millington & Quirk, 1960). Models specifically developed for repacked soils are also available (e.g., Moldrup et al., 2000). Moldrup et al. (2013) further developed the structure-dependent model, which conveniently represented both intact and repacked soils using an adjustable model parameter. The formulations of the above models are given in Table 2.

2.3.2 | Aggregated (two-region) soils

A limited number of models, representing both inter-aggregate (Region 1) and intra-aggregate (Region 2) regions in well-structured (aggregated) soils are also available (e.g.,

TABLE 2 Performance of selected classical and newly developed Two-region model against the measured D_p/D_o data expressed in terms of RMSE and BIAS. Soils are pasture soil at the University of Peradeniya, Sri Lanka (PD-P) and an arable soil at Meewathura, University of Peradeniya, Sri Lanka (PD-C)

Model	Equation	PD-P $d = 1.0 \text{ g cm}^{-3}$		PD-C (0-5 cm) $d = 1.42 \text{ g cm}^{-3}$		PD-C (5-10 cm) $d = 1.26 \text{ g cm}^{-3}$		PD-C (10-15 cm) $d = 1.23 \text{ g cm}^{-3}$	
		RMSE	BIAS	RMSE	BIAS	RMSE	BIAS	RMSE	BIAS
Buckingham (1904)	$\frac{D_p}{D_o} = \varepsilon^2$	0.0437	-0.0290	0.0594	-0.0468	0.0337	-0.0207	0.04095	-0.0250
Penman (1940)	$\frac{D_p}{D_o} = 0.66\varepsilon$	0.0534	0.0462	0.0418	0.0365	0.0716	0.0660	0.0781	0.0623
Marshall (1959)	$\frac{D_p}{D_o} = \varepsilon^{3/2}$	0.0454	0.0324	0.0293	0.0127	0.0552	0.0483	0.0529	0.0447
Millington (1959)	$\frac{D_p}{D_o} = \varepsilon^{4/3}$	0.0738	0.0615	0.0522	0.0421	0.0885	0.0809	0.0838	0.0777
Millington & Quirk (1960)	$\frac{D_p}{D_o} = \frac{\varepsilon^2}{\theta^{2/3}}$	0.0569	0.0244	0.0392	-0.0014	0.0482	0.0336	0.0354	0.0296
Millington & Quirk (1961)	$\frac{D_p}{D_o} = \frac{\varepsilon^{10/3}}{\theta^2}$	0.0755	-0.0133	0.0697	-0.0468	0.0462	-0.0218	0.0378	-0.0252
WLR-Marshall	$\frac{D_p}{D_o} = \varepsilon^{1.5} \left(\frac{\varepsilon}{\theta}\right)$	0.0504	-0.0072	0.0525	-0.0337	0.0307	-0.0048	0.0239	-0.0088
SWLR	$\frac{D_p}{D_o} = \varepsilon^{(1+C_m\theta)} \left(\frac{\varepsilon}{\theta}\right)$	0.0499	-0.0122	0.0545	-0.0367	0.0747	-0.0614	0.0870	-0.0663
Two-region model	$\frac{D_p}{D_o} = \frac{\alpha_1}{w^{\beta_1}} \left(\frac{\varepsilon}{\theta}\right)^{\beta_1} \varepsilon \leq w\theta$								
	$\frac{D_p}{D_o} = \alpha_1 + \frac{\alpha_2}{(1-w\theta)^{\beta_2}} \left(\frac{\varepsilon-w\theta}{\theta}\right)^{\beta_2} \varepsilon \leq (1-w)\theta$	0.0162	-0.0002	0.0106	-0.0004	0.0187	0.0000	0.0145	-0.0001

Resurreccion et al., 2007, 2008b, 2010). Theoretically based two-region models have also been proposed in recent literature (e.g., Ghanbarian et al., 2014) with limited validation across different soil structures. The two-region models typically assume that the two regions are analogous with respect to gas diffusivity. Some models, however, presume a non-linear behavior in Region 1 and a linear behavior in Region 2. A weighting factor, w , is usually included in the model to numerically distinguish the two-regions and is often used as a fitting parameter together with other model parameters.

In this study, we use a new two-region model which can be written in the form of:

For Region 1

$$\frac{D_p}{D_0} = \frac{\alpha_1}{w^{\beta_1}} \left(\frac{\varepsilon}{\emptyset} \right)^{\beta_1} \quad \varepsilon \leq w\emptyset \quad (3)$$

For Region 2

$$\frac{D_p}{D_0} = \frac{D_p}{D_0} \Big|_{\alpha=w\emptyset} + \frac{\alpha_2}{(1-w)^{\beta_2}} \left(\frac{\varepsilon - w\emptyset}{\emptyset} \right)^{\beta_2} \quad \varepsilon \leq (1-w)\emptyset \quad (4)$$

$$\frac{D_p}{D_0} = \alpha_1 + \frac{\alpha_2}{(1-w)^{\beta_2}} \left(\frac{\varepsilon - w\emptyset}{\emptyset} \right)^{\beta_2} \quad \varepsilon \leq (1-w)\emptyset \quad (5)$$

where \emptyset ($\text{cm}^3 \text{ cm}^{-3}$) is total porosity, α_1 , α_2 (dimensionless) are model scaling factors representing Region 1 and Region 2, respectively, while β_1 , β_2 (dimensionless) are corresponding shape factors. $\frac{D_p}{D_0} \Big|_{\alpha=w\emptyset}$ is the predicted gas diffusivity at $\varepsilon = w\emptyset$, which denotes the diffusivity when the inter-aggregate pores are completely dry and intra-aggregate pores are yet to be drained. Despite two equations for the Region 1 (Equation 3) and Region 2 (Equations 4 and 5), respectively, optimization is performed in one step for all fitting parameters.

2.3.3 | Tortuosity calculations

The tortuosity of the functional gaseous phase was calculated from measured D_p/D_0 and air-filled porosity data following Ball (1981) where tortuosity (τ) was defined as $\tau = \sqrt{\frac{\varepsilon}{\frac{D_p}{D_0}}}$.

We note here that the given equation defines the tortuosity as the ratio of the distance traversed by a gas molecule between two known points in the soil to the shortest (Euclidian) distance between the two points.

2.4 | Statistical Analysis

The conformity of the proposed model with the measured soil-gas diffusivity data and the overall performance of the existing models were evaluated and compared using RMSE and bias. To evaluate the overall fit of a model to the measured data, the RMSE was used:

$$\text{RMSE} = \sqrt{\frac{1}{n} \sum_{i=1}^n (d_i)^2} \quad (6)$$

and the bias was used to evaluate whether a model over-estimated (positive bias) or under-estimated the observations (negative bias).

$$\text{BIAS} = \frac{1}{n} \sum_{i=1}^n (d_i) \quad (7)$$

where d_i is the difference between the observed and predicted values, and n is the number of diffusivity measurements in a data set.

The correlation between the measured and predicted values were examined using the Pearson correlation coefficient (r).

3 | RESULTS AND DISCUSSION

3.1 | Particle size distribution

The measured particle size distributions of the two soils were successfully parameterized using the extended Rosin Rammler (1933) two-region particle size distribution function (Figure 1). In cultivated soils (PD-C), the difference in particle size distribution (PSD) between the different soil layers was not significant ($r > 0.99$), likely due to the frequent mixing as a result of tillage. Thus, the frequent tilling of cultivated soils created a homogenized soil layer (0–15 cm). Hence, the PSD data for the three cultivated soil depths were combined for numerical characterization. Importantly, both the soils showed a bimodal behavior with coarse fractions (w) equaling 0.89 and 0.80 for the PD-P and PD-C soils, respectively. The mean particle size of the pasture soil ($D_{50} = 0.24$ mm) was nearly two-fold larger than that for the cultivated soil ($D_{50} = 0.11$ mm), suggesting a marked difference in soil particle arrangements and hence distinct particle and pore network configurations. This, in turn, affects gas migration due to the difference in capillary-held water at differently sized pore regions.

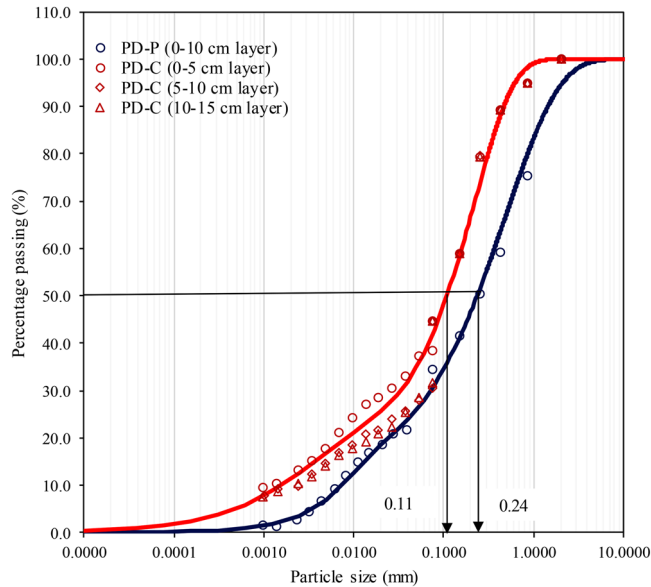


FIGURE 1 Measured particle size distribution for PD-P, PD-C soils of 0–5, 5–10, and 10–15 cm with the fitted curves for PD-P and PD-C according to extended Rosin Rammler (1933) two-region particle size distribution equation. For PD-P, $D_{50} = 0.24$, $\mu_c = 0.5000$, $\sigma_c = 0.740$, $\mu_f = 0.009$, $\sigma_f = 1.400$ and for PD-C, $D_{50} = 0.11$, $\mu_c = 0.225$, $\sigma_c = 1.007$, $\mu_f = 0.003$, $\sigma_f = 0.725$. PD-P, pasture soil at the University of Peradeniya, Sri Lanka; PD-C, arable soil at Meewathura, University of Peradeniya, Sri Lanka

3.2 | Soil-gas diffusivity

3.2.1 | Comparison of Taylor (1949) vs Currie (1960) methods for calculations

Observations and estimated D_p/D_o data calculated from either the Currie (1960) or Taylor (1949) methods are compared in Figure 2. Data for the Nishi-Tokyo cultivated and pasture soils are included in the analysis to strengthen the data analysis. Diffusivity data, measured over a broad range of air-filled porosity ($0\text{--}0.75\text{ cm}^3\text{ cm}^{-3}$), are presented across 15 different air-filled porosity zones (denoted by colours). Clearly, both methods yielded comparable D_p/D_o values within the air-filled porosity range $\leq 0.25\text{ cm}^3\text{ cm}^{-3}$. As the air-filled porosity increased above $0.25\text{ cm}^3\text{ cm}^{-3}$, the Currie (1960) method systematically over-estimated D_p/D_o with respect to the Taylor (1949) method, nearly reaching a 10% deviation at an air-filled porosity of $0.75\text{ cm}^3\text{ cm}^{-3}$. In general, a 5–10% disparity in D_p/D_o estimates was observed in aggregated soils above air-filled porosity of $0.25\text{ cm}^3\text{ cm}^{-3}$. The results corroborate well with previous observations, for example, Chamindu Deepagoda and Elberling (2015) reported a similar deviation (10–15%) for highly porous media, which they attributed to the absence of gas storage in Taylor’s method. As noted before, the Currie (1960) method, through the continuity equation coupled in Fick’s

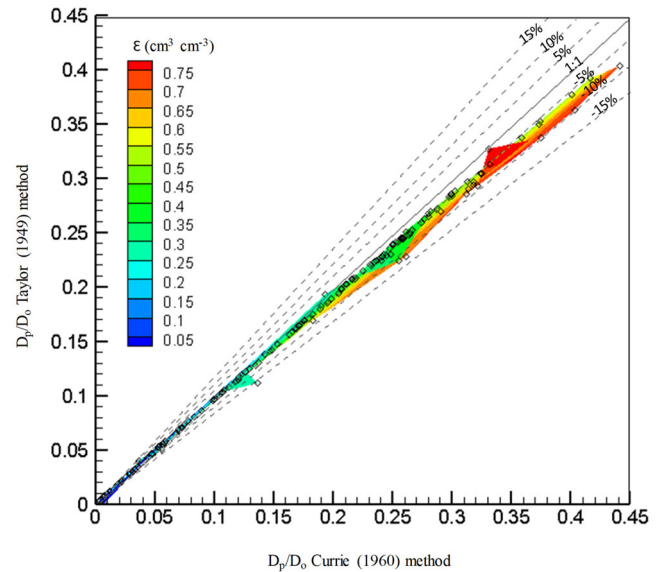


FIGURE 2 Tecplot comparison for Taylor (1949) method vs Currie (1960) method. Scatter points show the D_p/D_o for PD-P, PD-C, NT-P and NT-C soils. Variation of air-filled porosity (ϵ) is shown with flooded colors. The 1:1 line (solid line) and the \pm deviation for 5, 10, and 15% are given in dashed lines. PD-P, pasture soil at the University of Peradeniya, Sri Lanka; PD-C, arable soil at Meewathura, University of Peradeniya, Sri Lanka; NT-C, a Nishi-Tokyo cultivated soil (NT-C) from Tokyo, Japan; NT-P, a Nishi-Tokyo pasture soil from Tokyo, Japan

second law, considers gas storage in the D_p/D_o calculations whereas the Taylor (1949) equation is based only on Fick’s first law which does not take gas consumption, production or storage into account. As Rolston and Moldrup (2002) noted, the Currie (1960) method gives the ‘true’ D_p/D_o result and, by disregarding the gas storage, the Taylor (1949) method underestimates the diffusivity. Further, the Taylor (1949) method is not suitable for small time steps as the early phase concentration gradient of the experiment cannot be found with $(C_g - C_s)/L$ (where C_g and C_s [g gas m^{-3} air] are the concentration in the chamber and the concentration at the upper end of the soil core in contact with the atmosphere and L [m soil] is the length of the soil core) and a correction for chamber and core sizes are needed if the chamber height is small when applying the Taylor (1949) method for diffusivity calculation. Nevertheless, the Taylor (1949) method has some comparative advantages over the Currie (1960) method. The Taylor (1949) method is computationally less expensive since it does not require any iterative procedures or reference tables to calculate D_p/D_o . Air-filled porosity is not a calculation parameter for the Taylor (1949) method which enables calculations to be progressed at each step rather than waiting till the end to measure the air-filled porosity from oven-dried samples (Chamindu Deepagoda & Elberling, 2015). Thus, given the comparison of the two methods (Figure 2) we support the use of the Currie (1961) method for precise

calculation of D_p/D_o in bimodal agricultural soils with total porosities not exceeding $0.75 \text{ cm}^3 \text{ cm}^{-3}$, particularly under partially saturated conditions of higher gas storage in their inter-aggregate as well as intra-aggregate pores in contrast to unimodal soils having only inter-aggregate pores for gas storage.

3.2.2 | Modelling soil-gas diffusivity: A two-region model

The proposed two-region model (Equation 5) for characterizing soil gas diffusivity, for well-structured aggregated soil, was fitted to the measured diffusivity and data are shown in Figure 3, with diffusivity results for (a) pasture soils and (b) cultivated soils representing Sri Lankan and Japanese (Nishi-Tokyo) soils at different density levels. The parameterized two-region model is also shown in (a) and (b). Note that to capture the apparent linearity in the intra-aggregate pore region, β_2 was set equal to 1. Figure 3 clearly demonstrates unique soil structural fingerprints for D_p/D_o measurements in well-aggregated soils, as also noted in the literature (Resurreccion et al., 2008b). Notably in pasture soils (Figure 3a), with increasing compaction (from 1.0 g cm^{-3} to 1.3 g cm^{-3}), the boundary that demarcates the inter- and intra-aggregate pore regions shifted towards the left, implying a decrease in inter-aggregate pores (Φ_1) and an increase in intra-aggregate pores (Φ_2). This is because the

densification generally decreases the density of large, inter-aggregate pores while increasing the small, intra-aggregate pores (Currie, 1984). Currie further noted the complete disappearance of the inter-aggregate region at high density (1.29 g cm^{-3}). In the cultivated soils, however, the change from inter- and intra-aggregate porosity was less distinct for different soil depths. Further, density showed a reverse gradient with high-density soil at the top layer and the low-density soil at the bottom, as opposed to the pasture soils. This contrasting behavior in the cultivated soils is directly attributable to the frequent tilling operation, resulting in contrasting diffusivity characteristics.

For a particular air-filled porosity, increasing bulk density of the repacked Sri Lankan soils resulted in increased D_p/D_o at low air-filled porosities (mainly in the inter-aggregate region). This is because at the same air-filled porosity, moisture content in a dense soil is lower compared to a less dense soil and the higher moisture contents in less dense soils create interconnected water films thereby causing higher water induced tortuosity and less gas diffusivity (Moldrup et al., 2005a, 2005b). Fujikawa and Miyazaki (2005) explained this phenomenon from a different perspective using the concept of 'ineffective pores', which decrease with increasing compaction thus increasing the gas diffusivity. However, at higher air-filled porosities, where the intra-aggregate pore region drains, the less dense repacked soils showed higher diffusivities with increasing air-filled porosity due to decreased water induced tortuosity (Currie, 1984;

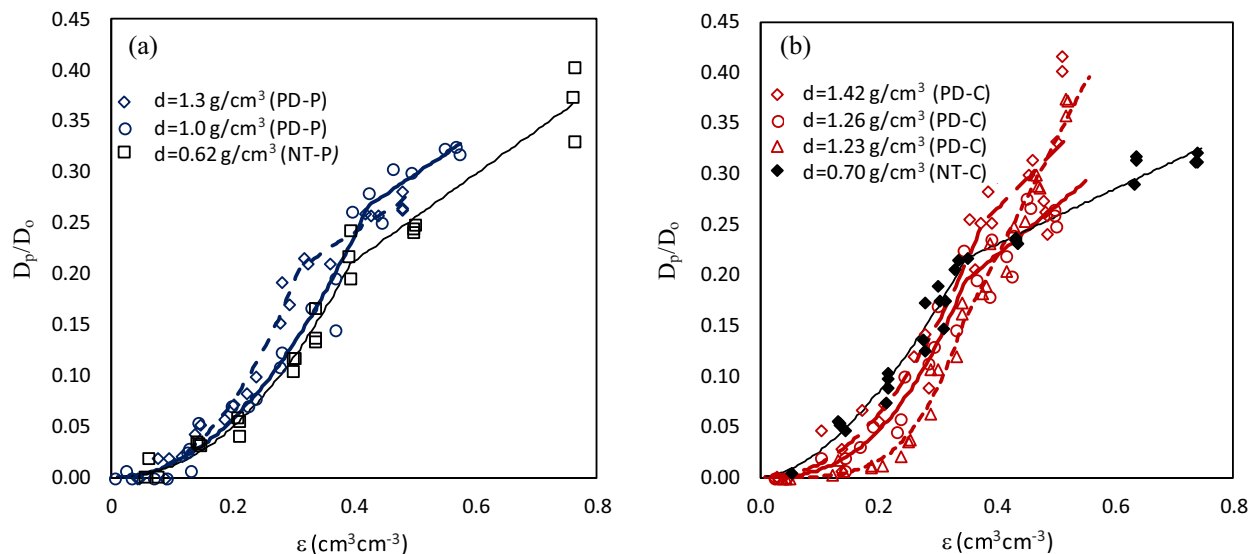


FIGURE 3 Scatter plot for the measured diffusivity versus soil air content. Panel (a) PD-P repacked samples of $d = 1.0 \text{ g cm}^{-3}$ and $d = 1.3 \text{ g cm}^{-3}$ along with their fitted curves; solid line for $d = 1.0 \text{ g cm}^{-3}$ and dashed line for $d = 1.3 \text{ g cm}^{-3}$, and NT-P repacked samples of $d = 0.62 \text{ g cm}^{-3}$ along with fitted curve. Panel (b) showing PD-C repacked samples of $d = 1.42 \text{ g cm}^{-3}$, $d = 1.26 \text{ g cm}^{-3}$ and $d = 1.23 \text{ g cm}^{-3}$, along with their fitted curves; long-dashed line for $d = 1.42 \text{ g cm}^{-3}$, solid line for $d = 1.26 \text{ g cm}^{-3}$ and short-dashed line for $d = 1.23 \text{ g cm}^{-3}$; and NT-C repacked samples of $d = 0.70 \text{ g cm}^{-3}$ along with the fitted curve for the newly developed Two-region model (thin black line). PD-P, pasture soil at the University of Peradeniya, Sri Lanka; PD-C, arable soil at Meewathura, University of Peradeniya, Sri Lanka; NT-C, a Nishi-Tokyo cultivated soil (NT-C) from Tokyo, Japan; NT-P, a Nishi-Tokyo pasture soil from Tokyo, Japan

Resurreccion et al., 2008a). Note that the gradients of the linear portions of the PD-P soils are almost similar for the two densities (0.39 and 0.41 for $d = 1.0 \text{ g cm}^{-3}$ and 1.3 g cm^{-3} , respectively). This can also be observed in the two top layers of the cultivated soils ($d = 1.46 \text{ g cm}^{-3}$ and 1.26 g cm^{-3}), but the bottom depth ($d = 1.23 \text{ g cm}^{-3}$) differed, due most likely to the mixing effect. The recorded gradients were 0.59 (for 1.0 g cm^{-3}), 0.48 (for 1.3 g cm^{-3}) and 1.15 (for $d = 1.23 \text{ g cm}^{-3}$). A similar behavior was also observed by Currie (1984), who noted that the alteration of intra-aggregate pore spaces was comparatively less with compaction. The results, therefore, confirm that the connectivity in the pore structure exerts control over the D_p/D_o . It is worth noting that the Nishi-Tokyo soils, despite their large difference with respect to soil type, showed similar behavior when compared to the Sri Lankan soils. The proposed two-region D_p/D_o model adequately parameterized the D_p/D_o of the selected agricultural soils distinguishing between their soil type and structure.

To demonstrate the promising behavior of the two-region model, we compared its performance with the classical and newly developed D_p/D_o models using scatterplot comparisons as shown in Figure 4. The statistical comparison of model performance for eight other models, based on the two statistical indices (Table 2), showed that the Buckingham (1904) and SWLR (Moldrup et al., 2013) models underestimated the results while the Penman (1940) and Marshall (1959) models overestimated the D_p/D_o measurements. The Millington and Quirk (1960, 1961) models markedly overestimated D_p/D_o at higher air-filled porosities and grossly under predicted D_p/D_o at low air-filled porosities, as typically observed in the literature. While the WRL-Marshall model (Moldrup et al., 2000), originally developed specifically for repacked soils, under predicted at lower air-filled porosities. Overall, the classical models lead to a marked bias of estimated values as compared to observations, probably due to the lack of provisions to capture the two-region characteristics. The two-region model (Equation 5), on the other hand, outperformed the classical models, yielding minimum RMSE and BIAS values.

3.3 | Tortuosity

The tortuosity values calculated using measured D_p/D_o data for the soils are shown in Figure 5, together with predictions from the two-region model as well as from the Buckingham (1904) and Penman (1940) models. As expected tortuosity values were high at high moisture contents due the water-induced pore tortuosity and disconnectivity, and this reduced markedly as the soils dried, leading towards the solid-induced tortuosity under drier conditions. In aggregated soils, the tortuosity values decrease as the inter-aggregate pores drain, reaching a minimum tortuosity when the inter-aggregate pore

space (Region 1) is completely drained. Further draining causes draining of intra-aggregate pore space, which opens up the more tortuous pore network within the aggregates, and thereby an increase in tortuosity values. This is particularly evident when the soil is strongly aggregated (Resurreccion et al., 2010), but is not distinctive in moderately aggregated soils, as observed in this study.

Compaction essentially decreases the larger pores and often results in an increase in the concentration of micropores in inter-aggregates, while decreasing the total inter-aggregate porosity. The compaction effect on intra-aggregate pore space, on the other hand, is not considerable (Resurreccion et al., 2008a), unless the compaction causes a breakdown in aggregate structure. Near saturation, the tortuosity is expected to be higher in less compacted soils, compared to compacted soils, due to the high moisture content and hence the moisture-induced tortuosity. However, as the soil drains, more dense soils may exhibit higher tortuosity than less dense soils, since dense soils tend to retain water in micropore-dominated pore structure. The tortuosity-air content relationship in aggregated soils thus provides another useful tool to fingerprint aggregated soils in relation to their state of aggregation (i.e., strongly aggregated, moderately aggregated, or weakly aggregated) as well as the level of compaction. The Penman (1940) model shows a constant value of tortuosity across the total air-filled porosity variation, typically yielding an upper-limit tortuosity. The Buckingham (1904) model, on the other hand, showed a nonlinear variation with decreasing tortuosity as the air-filled porosity increases. The developed two-region model also exhibited a nonlinear behavior with a sharp decline at high moisture contents while reaching a plateau at high air-filled porosities.

The two-region model developed based on the selected pasture and arable soils, and confirmed with agricultural soils from literature, provides a tool to more accurately estimate diffusion in soils influenced by aggregation. This in turn will improve models aiming to quantify the exchange of gases between the soil and the atmosphere, with respect to changes in land use management and practice, with implications for soil aggregation and climate change. This will be of significance for improved understanding of variations in O_2 diffusion and subsurface availability which, in turn, affects soil C storage (organic matter mineralization), CH_4 oxidation and N_2O emissions. While a better understanding of the aggregate effects on gas diffusivity within Region 1 potentially allows for the assessment of anaerobic and aerobic process dynamics that affect N_2O production, and/or further reduction to dinitrogen.

It should be mentioned herein that the results of this study were obtained under carefully controlled laboratory measurements in sieved and repacked samples representing one pasture soil and an arable soil. Further studies, including a wide range of pasture and arable soils, are needed to

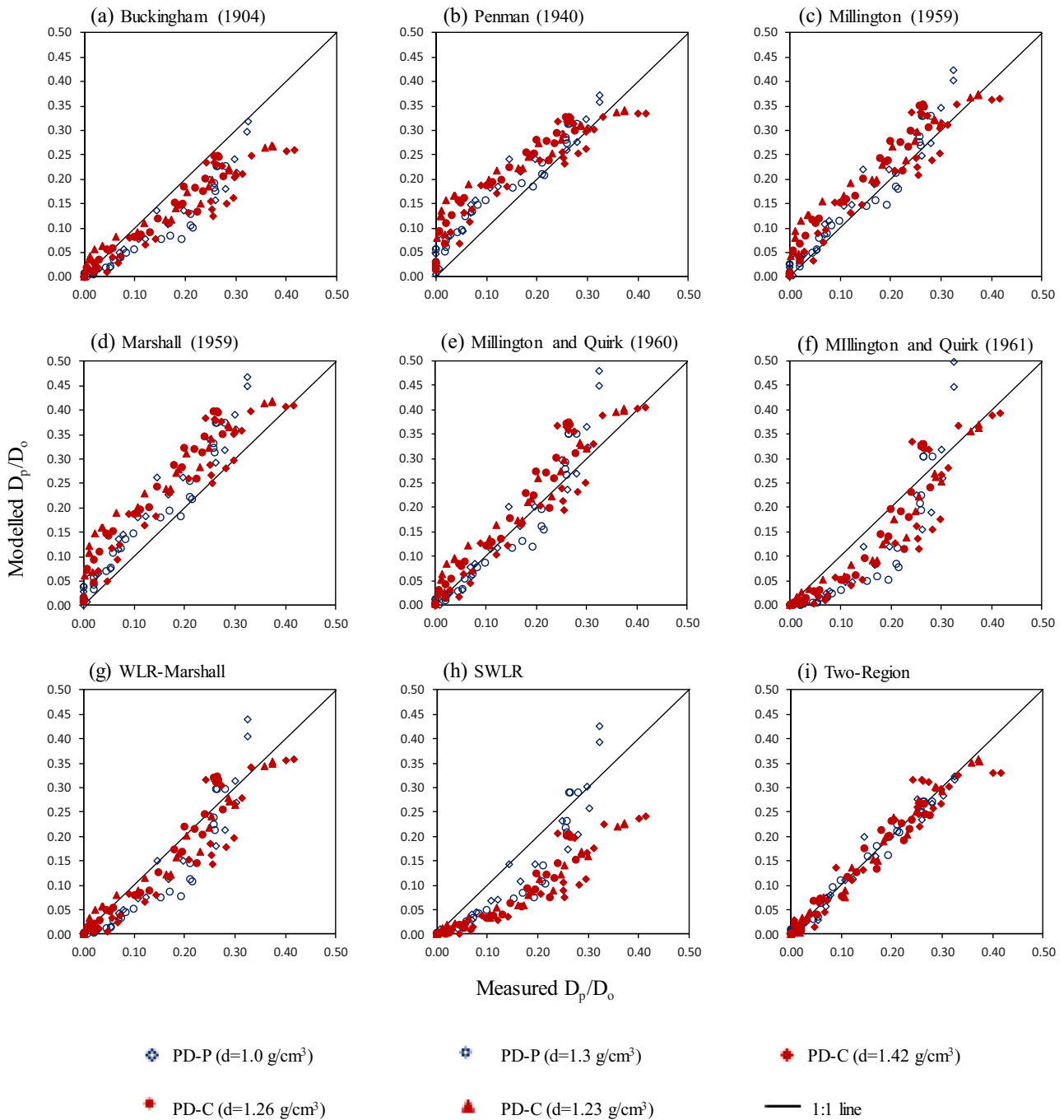


FIGURE 4 Scatter comparison of measured D_p/D_0 data points of PD-P $d = 1.0 \text{ g cm}^{-3}$, $d = 1.3 \text{ g cm}^{-3}$, and PD-C $d = 1.42 \text{ g cm}^{-3}$, $d = 1.26 \text{ g cm}^{-3}$, $d = 1.23 \text{ g cm}^{-3}$ against conventional models and recently developed models and the newly developed two-region model. The solid line shows the 1:1 relationship between the measured and modeled data. PD-P, pasture soil at the University of Peradeniya, Sri Lanka; PD-C, arable soil at Meewathura, University of Peradeniya, Sri Lanka

arrive at general conclusions on gas diffusivity behavior in selected soils. Further, soil density levels beyond the range $1.0\text{--}1.42 \text{ g cm}^{-3}$ for pasture soils and the densities in the pre-tilling cultivated soil were not considered in this study. Therefore, care should be taken when results of this study are compared with those from, for example, field experiments or intact samples where additional soil complexities (e.g.,

spatial structural and textural variability, variability in density) impose effects on results. We further emphasize that agricultural soil systems are continually dynamic in nature due to the factors such as grazing cycles, stocking rates, crop practices and soil management thus a long term analysis on diversified soils is needed in generalizing the applicability of the developed two-region D_p/D_0 model.

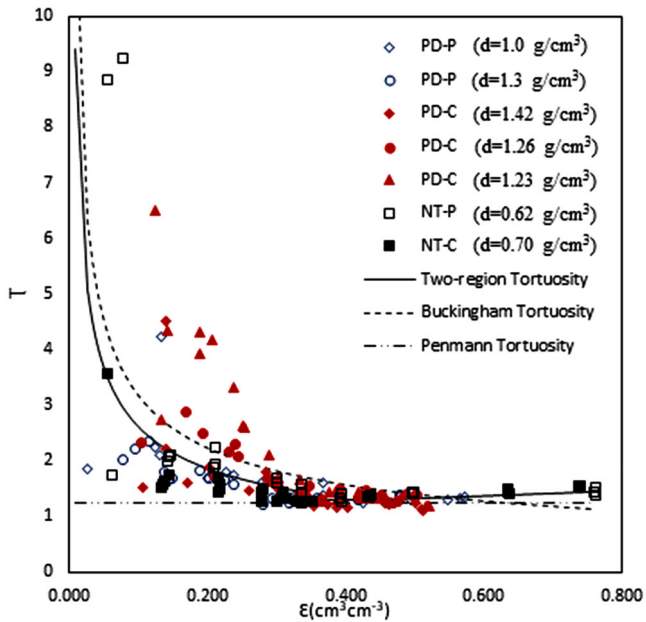


FIGURE 5 Scatter representation and fitting for tortuosity (T) variation against the air-filled porosity (ϵ) of PD-P $d = 1.0 \text{ g cm}^{-3}$ and $d = 1.3 \text{ g cm}^{-3}$, PD-C $d = 1.42 \text{ g cm}^{-3}$, $d = 1.26 \text{ g cm}^{-3}$, $d = 1.23 \text{ g cm}^{-3}$, NT-P $d = 0.62 \text{ g cm}^{-3}$, and NT-C $d = 0.70 \text{ g cm}^{-3}$. The fitted curve for two-region tortuosity, Buckingham tortuosity and Penman tortuosity are shown. PD-P, pasture soil at the University of Peradeniya, Sri Lanka; PD-C, arable soil at Meewathura, University of Peradeniya, Sri Lanka

4 | CONCLUSIONS

This study investigated the effect of soil structural status such as aggregation induced by compaction and tillage on soil gas diffusivity (D_p/D_o) in repacked pasture and cultivated soils sampled from Sri Lankan agricultural sites. We compared the two widely used methods of diffusivity calculation, Currie (1960) method and Taylor (1949) method, and observed a good agreement of results from the two methods at air-filled porosity below $0.25 \text{ cm}^3 \text{ cm}^{-3}$, but a 5–10% deviation above $0.25 \text{ cm}^3 \text{ cm}^{-3}$. The measured data were compared with eight recognized models for estimating soil diffusivity which yielded a marked disparity since none of them considered the distinct two-region characteristics of aggregated soils. The proposed multi-parameter two-region model accurately characterized and parameterized the measured D_p/D_o data and statistically outperformed the classic diffusivity models. The calculated gas phase tortuosity showed a strong nonlinear relationship with air-filled porosity and provided a good agreement with calculated tortuosity.

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