Density Effects on Soil Gas Diffusivity in Agricultural Soils

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***Abstract*—Accurate prediction of soil- gas diffusivity (D_p/D_o: where D_p and D_o are gas diffusion coefficients in soil and free air, respectively) and its variation with soil physical conditions (e.g., soil type/texture, soil density, soil moisture status) are important prerequisites for simulating subsurface gas migration and emission of greenhouse gases across soil-atmosphere continuum. Literature is abundant with studies using repacked soils for estimating soil-gas diffusivity, however they are unlikely to mimic realistic conditions in the subsurface, thereby leading to a marked mischaracterization of subsurface gas transport. In this study, measured soil-gas diffusivity in undisturbed soils sampled from differently characterized Danish soil profiles (total of 150 undisturbed soil samples) were used to investigate soil density effects on diffusive gas migration. The selected soils represent a wide range of natural and anthropogenic origins, including agricultural soils, forest soils, urban soils, landfill cover soils, etc. The measurements were within a selected range of matric potentials (−10 to −500 cm H_2O) typically representing natural field conditions in subsurface soil. The soils used for this study subjected to five different density categories (1.0-1.2, 1.2-1.4, 1.4-1.6, 1.6.-1.8, 1.8-2.0 g cm^{-3}) and showed peak diffusivity within the range of 1.4-1.6 g cm^{-3} as critical density window at a given suction. A series of predictive and descriptive gas diffusivity models were tested against the measured data for a model comparison. Results clearly distinguished the effect of soil structure status due to the accurate performance of SWLR model on measured diffusivity data.**

Keywords— soil density, soil gas diffusivity, soil types, Predictive models

I. Introduction

According to the Intergovernmental Panel on Climate Change (IPCC) [1], emission of greenhouse gases (primarily carbon dioxide, methane, and nitrous oxide) lead to significant regional and global climate changes. Although main greenhouse gas production occurs in natural systems, anthropogenic sources such as landfills, agricultural fields, and constructed wetlands also have contributed to an increased atmospheric presence [2]. As a high potent greenhouse gas, Mosier [3] has mentioned that CH_4 contributes nearly 25% of anticipated global warming, nearly one-third of which occurs in terrestrial ecosystems [4]. Among anthropogenic sources, landfills are responsible for approximately 7 - 20% of CH_4 emissions [5]. The gas migration through soil system is linked to soil physical properties such as soil texture and soil structure (e.g. soil density/compaction) and is mainly controlled by the physical, chemical, and biological processes in unsaturated soil layers. Subsurface migration of gases through the soil air phase and their subsequent emission across the soil–atmosphere interface occurs predominantly by diffusion and, to a lesser degree, by near-surface pressure fluctuations as explained by Penman [6] and Poulsen et al. [5], respectively.

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Soil gas diffusivity (the ratio of gas diffusion coefficients in soil and free air, D_p/D_o) is the key parameter which describes the diffusive transport of gases in partially saturated soils. Since, measurement of D_p/D_o is complicated to perform and instrumentally challenging in situ with sufficient control of the initial and boundary conditions (Rolston et al. [7]; Rolston and Moldrup [8]; Werner et al. [9]), descriptive/predictive models are frequently used to estimate D_p/D_o values from easily measurable parameters such as air-filled porosity and total porosity. Notwithstanding the presence of a wide range of predictive models, soil density and its associated changes to soil structure have been sparsely investigated in literature, although some general studies about effects of soil density on soil aeration are available. Buckingham [10], Stepniewski [11], Currie [12], Xu [13], Shimamura [14], Fujikawa and Miyazaki [15] have examined the direct effect of soil density on the gas transport parameters. According to the studies by Stepniewski [11] and Xu [13] on gas diffusion in differently textured soils, the effect of bulk density on the relationship between D_p/D_o and ε was less and have showed contradictory conclusions. Moreover, the observation by Fujikawa and Miyazaki [15] and Hamamoto [16] at a given air-filled porosity ε, D_p/D_o increased with the increment of bulk density. As explained by Currie [12], there was not any single relationship which can describe the changes in D_p/D_o with ε at different bulk densities. In this study, density effects on soil gas diffusivity were examined using undisturbed soils with different levels of density/compactness sampled across Denmark representing urban, agricultural, forest sites and as well as a final landfill cover soil.

II. Materials and Methods

A. Soils, sampling sites, and data

In this study, total of 150 literature data on undisturbed soil samples were considered. Soil samples were taken from eight different locations across Denmark, representing a wide range of soil texture, total porosities, and horizons. Undisturbed soil samples were collected using 100 cm^3 annular cores with 0.061 m internal dia., and 0.034 m length. Care was taken to ensuring minimum disturbance during sampling by driving the sharpened edge of annular core into the soil by means of a hammer. To prevent preferential air flow through the annular gap between the core and the sample, the end surfaces were trimmed, and the edges were kneaded with a knife. After that, the samples were end-capped and stored at 2°C. In the following text, soils are referred to as their sampling location (Skellingsted, Hjørring, Rønhave, Foulum, Jyndevad, Mammen, Gjorslev, and Poulstrup).

Urban soils were sampled at Skelingsted site which was located adjacent to an unlined municipal landfill operated as a dump of municipal solid and industrial waste from 1971 to 1990. According to Christophersen and Kjeldsen [17], the landfill was covered with 80 cm of sand and 20 cm of topsoil at the final closure and soil samples were collected at 70 cm depth. Hjørring also represents an urban soil which were sampled from a deep vadose zone profile from 4 to 5 m and 6 to 7 m depths at a former municipal gas work site. Both gas diffusivity data for Skellingsted and for Hjørring were partly presented by Poulsen [5] and Moldrup [18].

Under agricultural soils, Mammen and Gjorslev agricultural field soils and three lysimeter soils (Rønhave, Foulum, and Jyndevad) were included from Kawamoto [19,20]. Three lysimeter soils were excavated from the three locations, soils were air dried, crumbled to aggregates < 20 mm, and then packed in the bins incrementally in 10 cm layers to the same dry bulk density as occurred in the field located at Aarhus University, the Faculty of Agricultural Sciences at Research Centre Foulum. For further details on management and treatment practices of soils before sampling and packing procedure Kawamoto [20] and Lamandé [21] were referred respectively. Notably, Mammens and Gjorslev agricultural soil sites have been in agricultural use for centuries. Forest soil data from two medium-organic sandy layers collected in a natural mixed hardwood forest at Poulstrup representing two depth intervals,10 to 15 cm depth and 15 to 20 cm depth (Kruse [22], Moldrup [23]). Details of each layer for selected soils are given in Table I.

The distribution of soil textural classes for all 150 soil samples is illustrated in Fig 1. This Figure indicates that, except one clay layer from Hjørring (at 4.10 m depth), almost all the soil samples/layers from eight different locations can be texturally characterized as sand or sandy soils.

B. Measurement methods

To obtain the desired soil matric potentials for all soil samples, the method proposed by Klute [25] was used as follows. First, 100 cm^3 undisturbed soil cores were saturated inside sand boxes and samples were drained to the intended

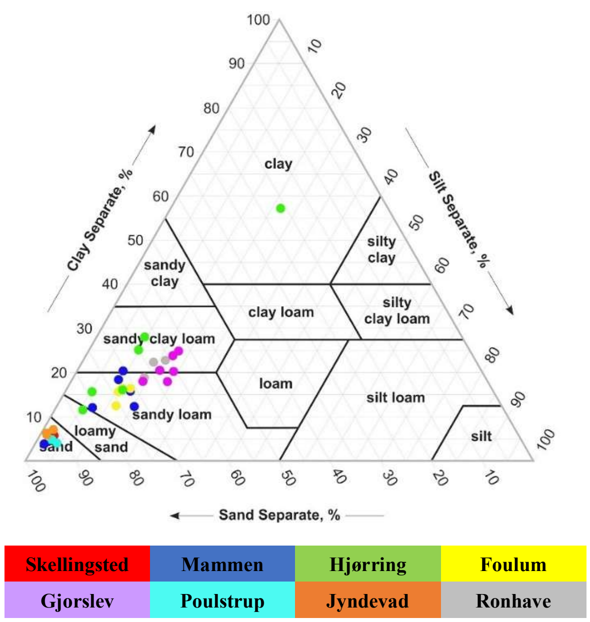


Fig. 1. Distribution of soil texture classes of 150 soil samples considered in this study

matric potential (ψ) using either hanging water columns or suction and pressure plate systems for $\psi > -100$ cm H_2O and for $\psi < -100$ cm H_2O, respectively. Matric potentials were selected in the range of -10 to -500 cm H_2O.

Using the one-chamber experimental setup initially presented by Taylor [26] and further developed by Schjønning [27], the values of D_p/D_o through soil samples were obtained. First, the chamber was flushed with 99.99% N_2 gas to make the chamber free of O_2. Then undisturbed soil core was placed on the chamber allowing atmospheric O_2 to diffuse through the soil sample into the chamber. Following the method outlined by Rolston and Moldrup [9], O_2 diffusion coefficient in soil (D_p) was calculated. As explained by Schjønning [28], time taken for each measurement differed due to the applied matric potential on soil sample and O_2 depletion due to microbial consumption can be neglected.

III. Statistical Analysis

To compare and test for general accuracy and tendency for overprediction or underprediction of existing models for gas diffusivity, two statistical indices were used as follows. RMSE was used to evaluate the model overall fit to the measured data.

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

Bias was used to evaluate whether a model over-estimated (positive bias) or under-estimated (negative bias) the observed data.

$$bias = \frac{1}{n}\sum_{i=1}^{n}(d_i) \qquad (2)$$

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

IV. Existing models from literature

Among the wide range of gas diffusivity models, the pioneering empirical model was introduced by Buckingham [11] as per (3).

$$\frac{D_p}{D_0} = \varepsilon^2 \qquad (3)$$

To prove this relationship, Buckingham used four different soils with varying moisture content and compactness, leading to conclude that gas diffusion in soils is not greatly affected by soil type. A series of single- parameter models were developed later by Penman [6]; Marshall [29]; Millington [30] in the given order. Later, by accounting density effects (implicitly) and soil type with the total porosity, Milington and Quirk [31,32] models were developed as soil type dependent models. Milington and Quirk 1960 [31] model is shown by (4):

$$\frac{D_p}{D_0} = \frac{\varepsilon^2}{\Phi^{2/3}} \qquad (4)$$

As per (5), Milington and Quirk 1961 [32] model is shown below:

TABLE I. SAMPLING LOCATIONS, DEPTHS, AND SOIL PHYSICAL CHARACTERISTICS

Location	Depth (m)	Texture	Clay %	Silt %	Sand %	Organic matter %	Total porosity #	Reference
Skellingsted	0.70	Sand	5.1	2.0	92.9	1.7	0.359 (0.020)	Poulsen [5]
Hjørring	4.00-4.50	Sandy clay loam	24.8	9.2	65.9	0.2	0.449 (0.040)	Moldrup [18]
Hjørring	4.10	Clay	56.6	21.0	22.3	0.2	0.502	Moldrup [18]
Hjørring	4.50-5.00	Sandy clay loam	26.9	9.2	63.9	0.2	0.456 (0.032)	Moldrup [18]
Hjørring	6.00-6.50	Sandy loam	15.7	10.8	73.4	2.1	0.382 (0.042)	Moldrup [18]
Hjørring	6.50-7.00	Loamy sand	11.2	5.0	83.8	1.6	0.404 (0.052)	Moldrup [18]
Gjorslev	0.05-0.25	Sandy clay loam	17.4	18.6	64.1	2.6	0.378 (0.013)	Kawamoto [19,20]
Gjorslev	0.33-0.53	Sandy clay loam	17.2	14.1	68.7	0.3	0.369 (0.008)	Kawamoto [19,20]
Gjorslev	0.80-1.00	Sandy clay loam	19.3	19.1	61.6	0.2	0.338 (0.013)	Kawamoto [19,20]
Gjorslev	2.05-2.25	Sandy clay loam	24.1	17.3	58.6	0.2	0.321 (0.006)	Kawamoto [19,20]
Gjorslev	3.50-3.70	Sandy clay loam	22.8	17.0	60.1	0.3	0.291 (0.008)	Kawamoto [19,20]
Gjorslev	4.65-4.85	Sandy clay loam	19.7	15.6	64.7	0.4	0.306 (0.037)	Kawamoto [19,20]
Mammen	0.05-0.25	Sandy loam	11.6	14.8	73.6	3.4	0.435 (0.005)	Kawamoto [19,20]
Mammen	0.30-0.50	Sandy clay loam	15.2	12.4	72.4	0.4	0.347 (0.013)	Kawamoto [19,20]
Mammen	1.10-1.30	Sandy clay loam	19.5	9.0	71.5	0.1	0.322 (0.005)	Kawamoto [19,20]
Mammen	2.05-2.15	Sandy clay loam	17.9	8.6	73.5	0.1	0.321 (0.010)	Kawamoto [19,20]
Mammen	3.40-3.60	Sandy loam	11.3	6.7	82.0	0.1	0.352 (0.010)	Kawamoto [19,20]
Mammen	5.40-5.60	Sand	3.6	0.9	95.5	0.0	0.389 (0.011)	Kawamoto [19,20]
Rønhave	0.00-0.30	Sandy clay loam	17.9	13.1	69.0	2.3	0.450 (0.025)	Kawamoto [19,20]
Rønhave	0.30-0.70	Sandy clay loam	21.7	13.5	64.8	0.5	0.436 (0.012)	Kawamoto [19,20]
Rønhave	0.70-1.40	Sandy clay loam	21.8	15.8	62.4	0.3	0.415 (0.010)	Kawamoto [19,20]
Foulum	0.00-0.30	Sandy loam	11.8	11.3	77.0	2.3	0.539 (0.020)	Kawamoto [19,20]
Foulum	0.30-0.60	Sandy loam	15.0	10.2	74.9	0.5	0.389 (0.017)	Kawamoto [19,20]
Foulum	0.60-0.90	Sandy clay loam	16.0	12.0	71.9	0.2	0.393 (0.002)	Kawamoto [19,20]
Foulum	0.90-1.40	Sandy clay loam	16.3	10.5	73.2	0.1	0.350 (0.005)	Kawamoto [19,20]
Jyndevad	0.00-0.30	Loamy sand	5.9	2.1	91.9	1.9	0.469 (0.019)	Kawamoto [19,20]
Jyndevad	0.30-0.70	Loamy sand	6.0	0.5	93.5	0.7	0.458 (0.010)	Kawamoto [19,20]
Jyndevad	0.70-1.40	Loamy sand	5.2	0.7	94.1	0.2	0.438 (0.013)	Kawamoto [19,20]
Poulstrup	0.10-0.15	Sand	3.7	3.1	93.2	3.7	0.519 (0.021)	Kruse [22]
Poulstrup	0.15-0.20	Sand	4.3	2.6	93.1	4.1	0.539 (0.031)	Moldrup [23]

Average values are given. Values in parentheses are standard deviations.
† Soil textures are classified based on the International Soil Science Society (ISSS) standard (Verheye and Ameryckx, [24])

$$\frac{D_p}{D_0} = \frac{\varepsilon^{\frac{10}{3}}}{\Phi^2} \quad (5)$$

Normally wet soils show large tortuosity for gas diffusion due to the generation of narrow pore throats between soil particles. Considering a water-induced linear reduction (WLR) of gas diffusivity in the presence of water, WLR–Marshall model (Moldrup [33]) was developed. The WLR model is expressed as (6):

$$\frac{D_p}{D_0} = \varepsilon^{1.5}\left(\frac{\varepsilon}{\Phi}\right) \quad (6)$$

With the lack of clear guidelines for model choice at a given soil state, the new structure-dependent WLR model (SWLR) was developed. By assuming a difference between the structureless soil state and the intact soil state, a porous media complexity factor, C_m, was introduced for the extension of WLR model. Using 290 soils representing well across soil depths, compaction, and texture, Moldrup [34] observed an excellent performance of SWLR model with $C_m = 2.1$ for gas diffusion of intact soils. As per (7), The SWLR model takes the form of:

$$\frac{D_p}{D_0} = \varepsilon^{(1+c_m\Phi)}\left(\frac{\varepsilon}{\Phi}\right) \quad (7)$$

V. RESULTS AND DISCUSSION

A. Effects of density and air-filled porosity

Fig. 2 shows measured D_p/D_o plotted against air filled porosity for all soil samples divided into five density categories in the ranges of 1.0-1.2, 1.2-1.4, 1.4-1.6, 1.6-1.8, 1.8-2.0 g cm^{-3}. As commonly observed D_p/D_o increased with the increase of air-filled porosity irrespective of the density. It can be seen that except for range E which had its data confined to the lower ε region (ε < 0.06), all the density categories had

D_p/D_o values that ranged across the entire ε with relatively little scatter. This behavior can be explained by agreeing well with Fujikawa and Miyazaki [15]. The increase in D_p/D_o with bulk density is due to the changes in the soil pore configuration (i.e., change in shape of soil pores) due to the natural compaction, resulting in a relative increase in the effective pore space. With the density increment of soils, volumetric solid content is increased with less water content, which creates less water bridging between particles and less water-induced tortuosity, leading to higher diffusivity values. As shown in Fig. 2, density category C gives the highest D_p/D_o values with the less effect of ineffective pore space. Thereafter D_p/D_o values again started to decrease due to less effective pore space since close packing and pore configuration.

Further to discuss the effect of bulk density, diffusivity data at two soil water matric potentials were selected. Fig. 3(a) and (b) illustrate the two dimensional graphical representations of soil gas diffusivity vs bulk density at suctions corresponding to pF = 1.7 (-50 cm H_2O) and 2.0 (-100 cm H_2O), respectively (note: pF = log |-Ψ, cm H_2O|).

Millington Quirk (1961), Buckinham (1904), WLR-Marshall, and SWLR predictive models for gas diffusivity were presented by solid lines. Maximum air filled porosity values at 1.40 g cm^{-3} bulk density for pF = 1.7 and 2.0 were used as reference values to generate predictive model lines. Selected models were able to capture the measured diffusivity values at middle range of density values, while overpredicting those at less and high densities. Note that the air-filled porosity variations (ε) across the bulk density are illustrated using 10 and 9 different air-filled porosities at pF = 1.7 and 2.0, respectively. Fig. 3(a) and (b) clearly demonstrate the presence of a density window which resulted a peak in gas diffusivity. This is against the generally expected tendency of decreasing gas diffusivity with increasing density. At a given suction, water is held in lower densities (1.1-1.3 g cm^{-3}) with high total porosity within larger pores creating additional water induced tortuosities for gas diffusion. Therefore, even at the same air-filled porosity, lower densities yielded lower diffusivities than higher densities due to the presence of high-water content surrounding the air-filled pores. At very high densities (1.6-1.8 g cm^{-3}) (with smaller total porosity), although the presence of water is less, the air-filled pore space is also less, and water is held in capillary-dominated smaller pores thus constraining the gaseous phase pore connectivity.

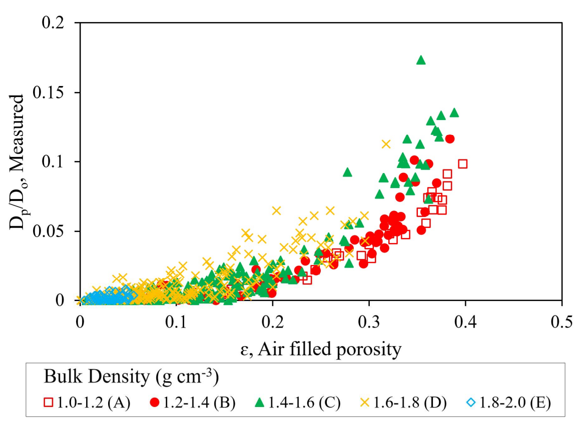


Fig. 2. Measured Gas diffusivities (D_p/D_0) against air - filled porosity (ε) for selected soils based on different density levels.

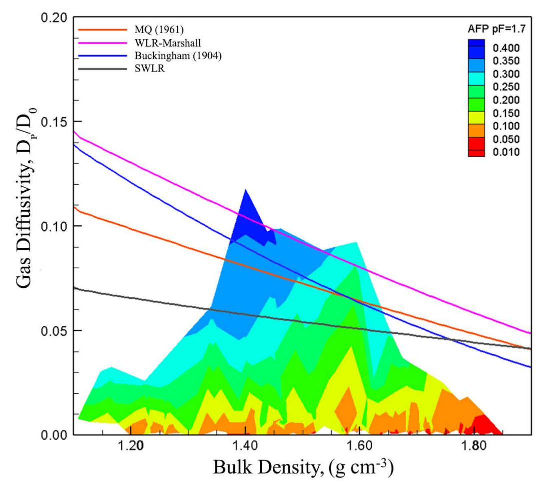
As a result, a diffusivity peak occurred at a medium-dense soil (1.4-1.6 g cm^{-3})

The observed peaks, however, did not occur under the same density for the two selected suction levels, implying that the peak location is moisture dependent. At higher suction (pF = 2.0), the D_p/D_o peaked at a higher density as compared to the peak occurred at the lower suction (pF = 1.7). This is due to the fact that more and more small-sized pores get drained at higher suction levels thus shifting the peak location towards the high-density direction.

B. Gas diffusivity model performances

The predicted gas diffusivity plotted against the measured soil gas diffusivity are shown in scatterplots in Fig. 4, using Buckingham (1904) (3), Millington Quirk (1961) (5), WLR - Marshall model (6), and SWLR model (7). Using RMSE (1) and bias (2) model performances were statistically evaluated. The detailed statistical analysis is given in Table II. According

(a)



(b)

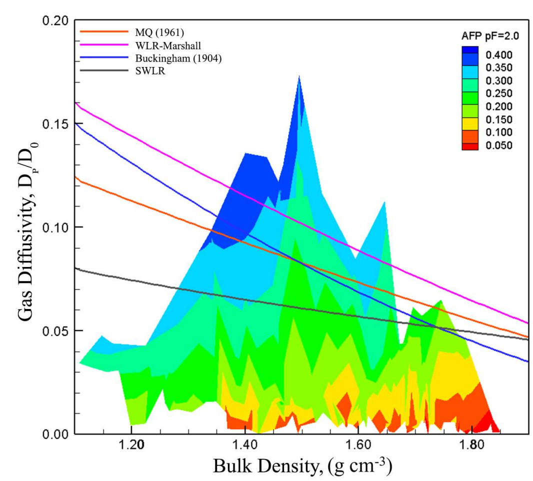


Fig. 3. Variation in soil gas diffusivity (D_p/D_o) for different bulk densities under different air-filled porosities (AFP) (a) at pF = 1.7 (-50 cm H_2O) soil water matric potential (b) at pF = 2.0 (-100 cm H_2O) soil water matric potential. The solid lines illustrate the predictions from selected gas diffusivity models.

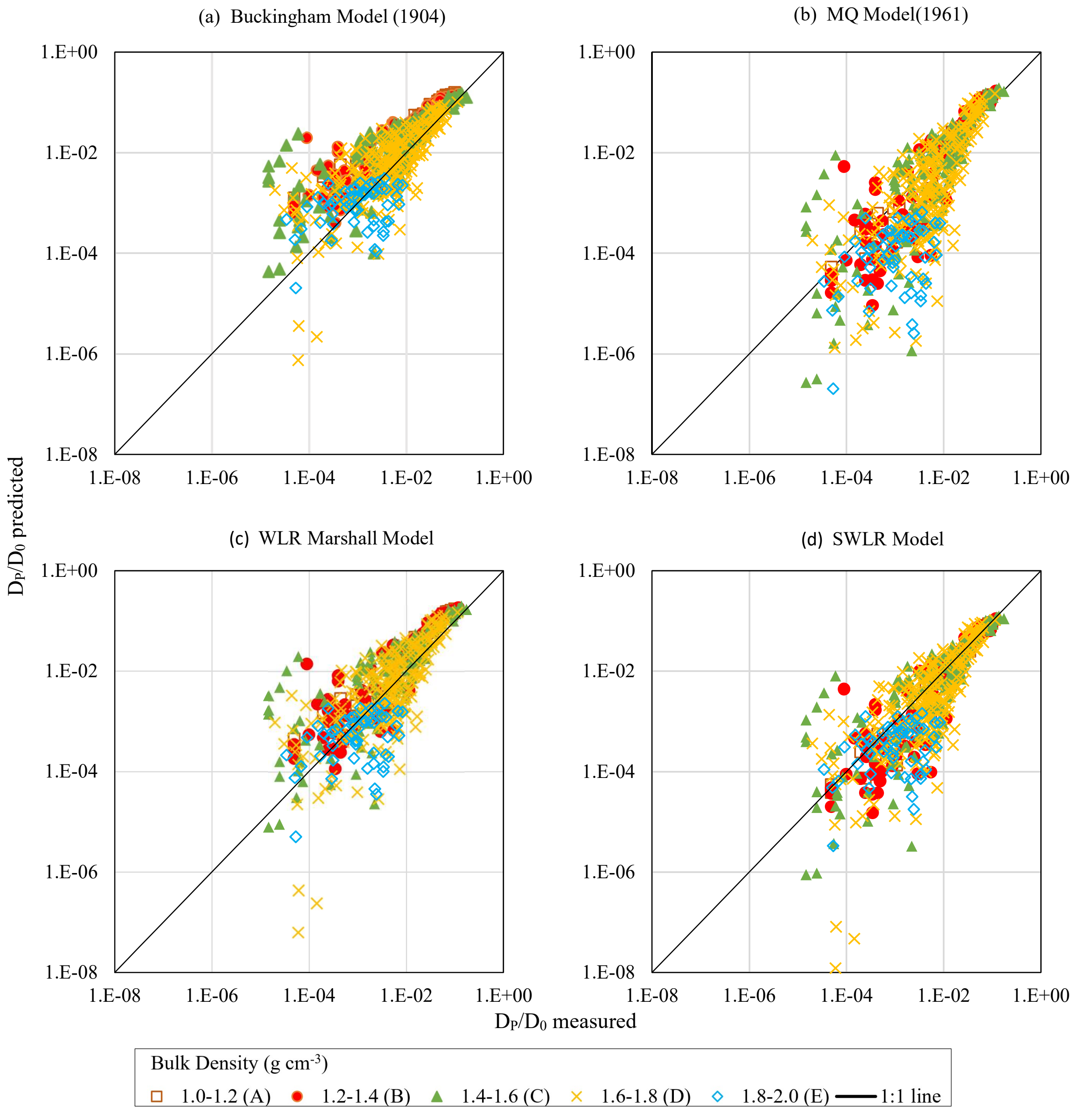


Fig. 4. Scatterplot comparison of measured and predictive D_p/D_o data points of A, B, C, D and E bulk density ranges for four existing models: (a) the Buckingham model (1904) (3), (b) Millington Quirk (1961) model (5), (c) the WLR-Marshall model (6), and (d) SWLR model (7)

to the statistical analysis WLR-Marshall (Moldrup [33]), indicates the weakest performance among the existing models with a significant overprediction. The Millington and Quirk (1961) model markedly overpredicted D_p/D_o at higher air-filled porosities and grossly underpredicted D_p/D_o at low air-filled porosities, as typically observed in the literature. Buckingham (1904), one of the earliest work on soil gas diffusivity, performed well on most of the soil than MQ (1961). Overall, above mentioned classical models (WLR-Marshall, MQ (1961), Buckingham) lead to a marked bias of estimated values as compared to observations, probably due to the lack of provisions to structure dependability of the soils. Notably, structure dependent water induce linear reduction (SWLR) model, outperformed the other classical models, yielding minimum RMSE and bias values as the best performed one to capture gas diffusivity behavior across soil texture and compaction levels accurately. Less-dense soils are better predicted than high dense soils by SWLR model. Normally at high densities, micropores are dominant and even at high suction levels water is held by capillary action. Due to this, water induced effects on diffusivity are high and leads to large scatter at high densities.

TABLE II. Performance of Selected Models Against the Measured D_p/D_o Data Expressed in Terms of RMSE and Bias

Model	Equation	RMSE	Bias
Buckingham (1904)	$D_p/D_o = \varepsilon^2$	0.0208	0.0109
Millington Quirk (1961)	$D_p/D_o = \varepsilon^{10/3}/\Phi^2$	0.0177	0.0056
WLR – Marshall	$D_p/D_o = \varepsilon^{1.5}(\varepsilon/\Phi)$	0.0268	0.0137
SWLR	$D_p/D_o = \varepsilon^{(1+Cm.\Phi)}(\varepsilon/\Phi)$	0.0083	-0.0003

VI. CONCLUSIONS

This study investigated the density effects on soil gas diffusivity of agricultural soils subjected to matric potentials between -10 cm to -500 cm H_2O as usual subsurface moisture conditions. Total 150 Danish soil samples were studied to evaluate the effect of density, air filled porosity, D_p/D_o and comparison of predictive model performances. Results identified a critical density window (1.4-1.6 g cm^{-3}) which resulted in a peak in D_p/D_o at a given suction status. The location of peak shifted to a higher density at higher suction level, and hence was moisture dependent. The measured data were compared with three existing models for estimating soil diffusivity which yielded a marked disparity since none of them considered the soil structure status. The SWLR model accurately characterized the measured D_p/D_o data and statistically outperformed the other three models. Overall, the results confirmed a pronounced effect of soil density on diffusive transport of gases in partially saturated soils and emphasized the need of selective model applications to accurately characterize soil density effects.

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