Comparison of Classical and Recent Predictive Models for Soil-Gas Diffusivity

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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 air-filled porosity (ε) is important for understanding soil aeration and subsurface greenhouse gas emissions and thereby to characterize essential soil functional services in terrestrial ecosystems. Since measuring D_p/D_o is instrumentally challenging and requires maintaining controlled boundary conditions, different predictive models have been developed to estimate D_p/D_o from easily measurable soil properties such as air-filled porosity (ε) and soil total porosity (Φ). In this study, a total of 593 gas diffusivity measurements conducted on 150 data from differently characterized undisturbed Danish soils were used to evaluate the performance of five prospective predictive models developed over the period of 1904 -2013. The selected soils represent agricultural soils, forest soils, urban soils, and landfill cover soils and measurements were within a selected range of matric potentials (−10 to −500 cm H_2O) typically occurring in subsurface. Results of the model comparison made using two statistical indices (RMSE and Bias) showed that widely used model for repacked soils made a significant overprediction of undisturbed data. This study clearly distinguished the effect of soil structure status on soil gas diffusivity as demonstrated by the best performance of SWLR model over the other predictive models by yielding minimum RMSE and bias.**

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

I. Introduction

Emission of greenhouse gases (GHG), primarily carbon dioxide (CO_2), methane (CH_4), and nitrous oxide (N_2O), has been attributed to cause significant regional and global climate changes [1]. Although main greenhouse gas production occurs in natural systems, anthropogenic sources such as landfills, agricultural fields, and constructed wetlands also contribute to the increased atmospheric abundance of GHG [2]. As a powerful greenhouse gas, CH_4 contributes nearly 25% of anticipated global warming [3], nearly one-third of which occurs in terrestrial ecosystems [4]. Landfills are responsible for approximately 7 - 20% of CH_4 emissions [5] from anthropogenic sources. To mitigate this excessive atmospheric presence of GHG, accurate prediction and modelling of gas movement in soil as related to varying soil physical properties under natural field conditions is essential.

Migration of gases in the subsurface occurs primarily by diffusion [6]. The uptake or emission of gases across the soil-atmosphere continuum is mainly controlled by diffusion, accompanied with further acceleration due to advection caused by near-surface pressure fluctuations [7]. Diffusive transport of gases in soils can be described by soil gas diffusivity (the ratio of gas diffusion coefficients in soil and free air, D_p/D_o). Since measuring D_p/D_o is complicated by the need of specific apparatus and controlled boundary conditions, predictive models, together with easily measurable parameters such as air-filled porosity and total porosity, are widely used as an alternative. Series of predictive models have successively appeared over the history and the Buckingham (1904), Penman (1940), Millington and Quirk (1960 and 1961), WLR–Marshall (2000), and Troeh (1982) are some notable models developed over the last century. Later models have attempted to account additional soil complexities (e.g., soil density and moisture-induced tortuosity) to better characterize soil structural impacts on soil gas diffusivity. Some notable recent models, among others, include Resurreccion et al. [8], Chamindu Deepagoda et al. [9,10], and Moldrup et al. [11].

In this study, the performance of a series of widely recognized soil-gas diffusivity models was reviewed using undisturbed soils with different levels of compactness, soil texture, horizons, and total porosities sampled across Denmark representing a wide range of natural and anthropogenic ecosystems.

II. Materials and Methods

A. *Soil Types and Data*

In this study, 150 literature data on undisturbed soil samples were considered. Soil samples have been collected from eight different locations across Denmark, representing a wide range of soil textures, total porosities, and horizons. In the following text, soils are referred according to the sampling location (Skellingsted, Hjørring, Rønhave, Foulum, Jyndevad, Mammen, Gjorslev, and Poulstrup). Annular cores with 100 cm^3 volume (0.061 m internal dia., and 0.034 m length) have been used for sampling at all locations with similar dimensions. During sampling, care has been taken to ensure minimum disturbance by driving the sharpened edge of the 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 have been trimmed, the edges have been kneaded with a knife. Before measurements, soil samples have been end-capped and kept at 2°C.

The 150 soil samples can be divided into two main categories: urban soils, and agricultural and forest soils. Urban soils have been collected at Skelingsted site which was located adjacent to an unlined municipal landfill. It has been operated as a dump of municipal solid waste and industrial waste from 1971 to 1990. The landfill has been covered with 80 cm of sand and 20 cm of topsoil at the final closure [12] and soils have been sampled at 70 cm depth. Hjørring also represents an urban soil, has been 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 have been partly presented by Poulsen et al. [7] and Moldrup et al. [13].

Two agricultural soils Mammen and Gjorslev, three lysimeter soils with different soil textures (Rønhave, Foulum, and Jyndevad) have been considered from Kawamoto et al. [14,15]. Mammens and Gjorslev agricultural soil sites have been in agricultural use for centuries. Three lysimeter soils have been excavated from three locations, air dried and have been crumbled to aggregates < 20 mm. After that aggregates have been 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 the management and treatment practices of the soils before sampling, and on the packing procedure, see Kawamoto et al. [15] and Lamandé et al. [16], respectively. Forest soils have been collected from two medium-organic sandy layers in a natural mixed hardwood forest at Poulstrup representing two depth intervals,10 to 15 cm [17] and 15 to 20 cm [18]. Sampling location, depths, texture, and soil physical characteristic details of selected soils are given in Table I.

B. *Measurement Methods*

Using the method proposed by Klute [20], the desired soil matric potentials for all soil samples have been obtained as follows. The undisturbed soil samples have been saturated inside the sand boxes and then drained to the intended 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 have been selected in the range of -10 to -500 cm H_2O. The values of D_p/D_o through soil samples have been obtained using the one-chamber experimental setup initially presented by Taylor [21] and further developed by Schjønning [22]. First, the chamber has been flushed with 99.99% N_2 gas to make the chamber free of O_2, and then the soil core has been placed on the chamber allowing atmospheric O_2 to diffuse through the soil sample into the chamber. The O_2 diffusion coefficient in soil (D_p) has been calculated using the method outlined by Rolston and Moldrup [23]. Time taken for each measurement differs due to the applied matric potential on soil sample and O_2 depletion due to microbial consumption has been neglected in this study.

III. Statistical Analysis

To evaluate the performance (overprediction or underprediction) of existing gas diffusivity models, two statistical indices were used as follows. RMSE was used to evaluate the model overall fit to the measured data.

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

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

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

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TABLE I. SOIL SAMPLING DETAILS 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 [7]
Hjørring	4.00-4.50	Sandy clay loam	24.8	9.2	65.9	0.2	0.449 (0.040)	Moldrup [13]
Hjørring	4.10	Clay	56.6	21.0	22.3	0.2	0.502	Moldrup [13]
Hjørring	4.50-5.00	Sandy clay loam	26.9	9.2	63.9	0.2	0.456 (0.032)	Moldrup [13]
Hjørring	6.00-6.50	Sandy loam	15.7	10.8	73.4	2.1	0.382 (0.042)	Moldrup [13]
Hjørring	6.50-7.00	Loamy sand	11.2	5.0	83.8	1.6	0.404 (0.052)	Moldrup [13]
Gjorslev	0.05-0.25	Sandy clay loam	17.4	18.6	64.1	2.6	0.378 (0.013)	Kawamoto [14,15]
Gjorslev	0.33-0.53	Sandy clay loam	17.2	14.1	68.7	0.3	0.369 (0.008)	Kawamoto [14,15]
Gjorslev	0.80-1.00	Sandy clay loam	19.3	19.1	61.6	0.2	0.338 (0.013)	Kawamoto [14,15]
Gjorslev	2.05-2.25	Sandy clay loam	24.1	17.3	58.6	0.2	0.321 (0.006)	Kawamoto [14,15]
Gjorslev	3.50-3.70	Sandy clay loam	22.8	17.0	60.1	0.3	0.291 (0.008)	Kawamoto [14,15]
Gjorslev	4.65-4.85	Sandy clay loam	19.7	15.6	64.7	0.4	0.306 (0.037)	Kawamoto [14,15]
Mammen	0.05-0.25	Sandy loam	11.6	14.8	73.6	3.4	0.435 (0.005)	Kawamoto [14,15]
Mammen	0.30-0.50	Sandy clay loam	15.2	12.4	72.4	0.4	0.347 (0.013)	Kawamoto [14,15]
Mammen	1.10-1.30	Sandy clay loam	19.5	9.0	71.5	0.1	0.322 (0.005)	Kawamoto [14,15]
Mammen	2.05-2.15	Sandy clay loam	17.9	8.6	73.5	0.1	0.321 (0.010)	Kawamoto [14,15]
Mammen	3.40-3.60	Sandy loam	11.3	6.7	82.0	0.1	0.352 (0.010)	Kawamoto [14,15]
Mammen	5.40-5.60	Sand	3.6	0.9	95.5	0.0	0.389 (0.011)	Kawamoto [14,15]
Rønhave	0.00-0.30	Sandy clay loam	17.9	13.1	69.0	2.3	0.450 (0.025)	Kawamoto [14,15]
Rønhave	0.30-0.70	Sandy clay loam	21.7	13.5	64.8	0.5	0.436 (0.012)	Kawamoto [14,15]
Rønhave	0.70-1.40	Sandy clay loam	21.8	15.8	62.4	0.3	0.415 (0.010)	Kawamoto [14,15]
Foulum	0.00-0.30	Sandy loam	11.8	11.3	77.0	2.3	0.539 (0.020)	Kawamoto [14,15]
Foulum	0.30-0.60	Sandy loam	15.0	10.2	74.9	0.5	0.389 (0.017)	Kawamoto [14,15]
Foulum	0.60-0.90	Sandy clay loam	16.0	12.0	71.9	0.2	0.393 (0.002)	Kawamoto [14,15]
Foulum	0.90-1.40	Sandy clay loam	16.3	10.5	73.2	0.1	0.350 (0.005)	Kawamoto [14,15]
Jyndevad	0.00-0.30	Loamy sand	5.9	2.1	91.9	1.9	0.469 (0.019)	Kawamoto [14,15]
Jyndevad	0.30-0.70	Loamy sand	6.0	0.5	93.5	0.7	0.458 (0.010)	Kawamoto [14,15]
Jyndevad	0.70-1.40	Loamy sand	5.2	0.7	94.1	0.2	0.438 (0.013)	Kawamoto [14,15]
Poulstrup	0.10-0.15	Sand	3.7	3.1	93.2	3.7	0.519 (0.021)	Kruse [17]
Poulstrup	0.15-0.20	Sand	4.3	2.6	93.1	4.1	0.539 (0.031)	Moldrup [18]

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, [19]

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. DIFFUSION STUDIES AND EXISTING MODELS

In 1904, the pioneering model was introduced by a U.S. soil physicist, Edgar Buckingham through his ground-breaking gas diffusion experiments. Buckingham carried out aeration and gas diffusion experiments in four different soils with varying moisture content and compactness. He calculated the gas diffusion coefficient, D_p, and found a close relation between D_p/D_o and soil air content, ε, as per (3) leading to conclude that gas diffusion in soils is not greatly affected by soil type [6].

$$\frac{D_p}{D_0} = \varepsilon^2 \tag{3}$$

A series of single parameter models were developed later by Penman [24]; Marshall [25]; Millington [26] in the given order until the next generation of models started to incorporate some soil type and density effects through the soil total porosity (Φ). R. J. Millington developed theoretically based equations together with another scientist, J. P. Quirk. The Millington and Quirk (1960) model [27] is shown by (4):

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

The Millington and Quirk (1961) model [28], the almost universally-accepted model adopted in most classical numerical tools for gas transport, can be presented in the form of (5) as follows:

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

In wet soils, water held at bottlenecks potentially create large tortuosity for gas diffusion. Moldrup et al. [29], considered this water blockage effect into account by assuming a water-induced linear reduction (WLR) for gas diffusivity, yielding the WLR–Marshall model [29] as given in (6) below:

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

Chamindu Deepagoda et al. [10] introduced the density corrected (D-C) model concept on intact soil data including higher-organic soils. Based on this study, they suggested that soil compaction more than soil type was the major control on gas diffusivity [10]. The D-C model can be written as (7):

$$\frac{D_P}{D_0} = 0.1\left[2\left(\frac{\varepsilon}{\Phi}\right)^3 + 0.04\left(\frac{\varepsilon}{\Phi}\right)\right] \quad (7)$$

Due to the lack of clear guidelines for model choice at a given soil state, by considering the difference between the repacked or structureless soil state and the intact soil state, the second version of WLR model was developed by Moldrup et al. [11] by introducing a porous media complexity factor C_m that is assumed to be related to soil density and thus total porosity. The new structure-dependent WLR model (SWLR) (8) was proposed with $C_m = 2.1$ for gas diffusion in intact soils.

$$\frac{D_p}{D_0} = \varepsilon^{(1+c_m\Phi)}\left(\frac{\varepsilon}{\Phi}\right) \quad (8)$$

V. Results and Discussion

Fig. 1 shows the scatterplot comparison of predicted gas diffusivity plotted against the measured soil gas diffusivity values for the Buckingham (1904) (3), Millington Quirk (1961) (5), WLR -Marshall model (6), D-C model (7), and SWLR model (8).

Model performances were statistically evaluated using RMSE (1) and bias (2) and a detailed statistical analysis is given in Table II. WLR-Marshall model indicates the weakest performance with a significant overprediction. The widely accepted MQ (1961) and Buckingham (1904) models markedly overpredicted D_p/D_o at higher air-filled porosities and grossly underpredicted D_p/D_o at low air-filled porosities. The empirically-based Buckingham (1904) model performed better on most of the soils over the theoretically-based MQ (1961).Notably, the semi-conceptual WLR-Marshall model, developed and validated for repacked soils, disregards the intrinsic cohesion among particles through chemical bonds in undisturbed soils, and hence shows higher predictive values for intact soils. Overall, Density-Corrected model (2011) seems a more accurate capture of gas diffusivity behavior than other models (except SWLR model) as it was developed for intact soils by considering density and moisture effect. Above mentioned classical models (Buckingham, WLR-Marshall, MQ (1961)) show significant bias, probably due to the lack of provisions for structure-induced complexity in intact the soils. Amongst all models, the SWLR model, with particular account on soil structural effects, showed the best performance by statistically outperforming all other models, thus yielding minimum RMSE and bias values.

VI. Conclusions

This study reviewed the performance of selected soil-gas diffusivity models using 150 undisturbed Danish soil samples subjected to matric potentials between -10 cm to -500 cm H_2O as naturally occuring subsurface moisture conditions. The measured data were compared with five existing models for estimating soil-gas diffusivity and four of them yielded a marked disparity since none of them adequately accounted the soil structure effects. Overall, results identified that SWLR model accurately predicted the measured D_p/D_o data and statistically outperformed the other four models with minimum RMSE and Bias. Thus, the SWLR model seems to be a useful model for both intact and repacked soils with its adaptable complexity factor (C_m), and hence can be a promising model to be incorporated in future gas transport-related numerical tools.

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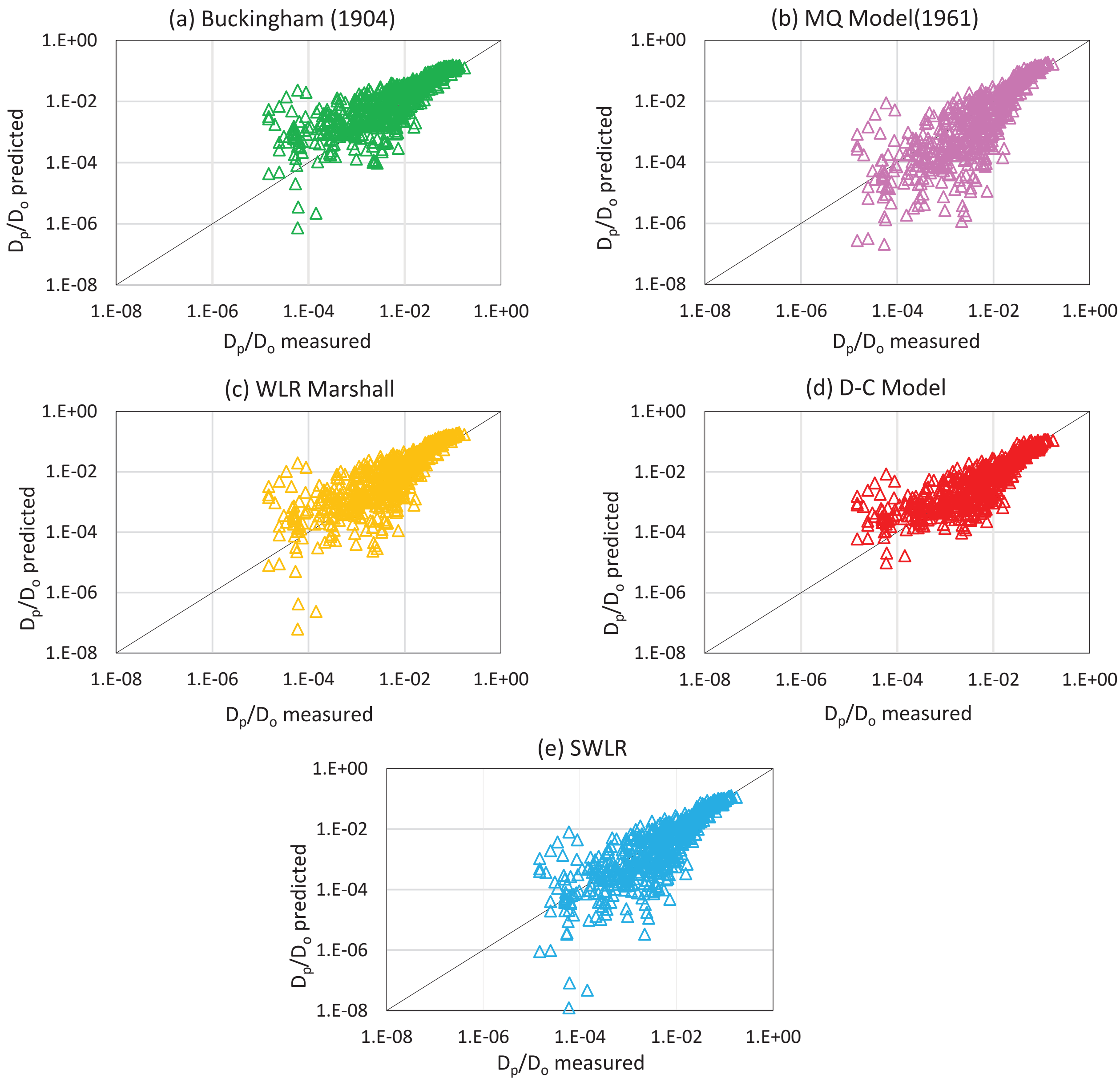

Fig. 1. Scatterplot comparison of measured and predictive D_p/D_o data points for selected models: (a) the Buckingham model (1904) (3), (b) Millington Quirk (1961) model (5), (c) the WLR-Marshall model (6), (d) D-C model (7), and (e) SWLR model (8)

TABLE II. Selected Gas Diffusivity Model Performances in terms of RMSE and Bias

Model	RMSE	Bias
Buckingham (1904)	0.0208	0.0109
MQ Model (1961)	0.0177	0.0056
WLR – Marshall	0.0268	0.0137
D-C Model	0.0093	-0.0006
SWLR	0.0083	-0.0003

It has to be mentioned, while this comparison was based on the literature data from various sources, the applicability of the results should be further verified with other classes of soils which representing wide range of structures, horizons, and densities.

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