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Review of Recent Progress in Predicting Gas Transport Parameters for Undisturbed Andisols : Campbell b Dependent Models for Gas Diffusivity and Air Permeability

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Abstract

This paper reviews and connects recent studies on gas transport parameter models for Japanese volcanic ash soils (Andisols). Soil water retention from -1 kPa to -1.5 MPa of matric potential for differently-textured, undisturbed Andisols from three prefectures in Japan was well described by the simple Campbell (1974) model. Gas diffusivity in the same matric potential range was well predicted by two recent soil type (Campbell *b*) dependent models, while the classical Millington and Quirk (1961) model markedly under-predicted gas diffusivity for all Andisols. Air permeability (k_a) in wet to medium moist soil (from -1 to -100 kPa of matric potential) was also well predicted by a Campbell *b* dependent model, provided that k_a at -10 kPa of matric potential was measured and used as a reference point in the model. In conclusion, Campbell-based models appear highly useful for describing pore characteristics and predicting gaseous phase transport parameters in Andisols.

Key words : gas diffusion, gas transport, Campbell soil water retention model, BBC gas diffusivity model, soil structure fingerprint

1. Introduction

Predictive models for the key transport parameters in the soil gaseous phase (the soil gas diffusion coefficient and the soil air permeability) have until recently only been tested within limited ranges of soil pore size distribution and soil total porosity (Moldrup *et al.*, 2001). Since volcanic ash soils (Andisols) exhibit soil physical properties that are quite different from normal (non-volcanic) mineral soils, including larger total porosities and higher soil water retention (broader pore size distributions), data for Andisols should prove highly valuable in testing the general validity of predictive models for the gas diffusion coefficient (gas diffusivity) and air permeability, both being functions of soil air-filled porosity and soil type. The distinctive soil physical characteristics of Andisols is typically caused by allophane, a non-crystaline and highly porous mineral. Andisols are found in all parts of Japan and cover more than 16% (more than 60,000 km^2) of the total land area of Japan (Adachi, 1971). For more on Andisol and allophane properties and characteristics, we refer to Henmi (1988), Shoji *et al.* (1993), Iwata *et al.* (1995), and So (1999).

Gas diffusivity (D_p) controls gas transport and fate in natural soil systems where diffusive gas transport is typically dominating compared with convective gas transport. Important examples are soil aeration (Buckingham, 1904) and its effects on plant health (Osozawa *et al.,* 1994, Hasegawa, 1994), the emission of fumigants at soil fumigation sites (Brown and Rolston, 1980), the diffusion and volatilisation of organic chemicals from polluted soils (Petersen *et al.,* 1996), and the diffusion and biodegradation of greenhouse gases such as methane and carbon dioxide (Kruse *et al.,* 1996, Yoshikawa and Hasegawa, 2000). One of the "rules of thumb" in soil physics is an almost universal use of the Millington and Quirk (MQ, 1961) equation to predict the changes in D_p with air-filled porosity (ε) in transport and fate models for gaseous compounds in soil. Since the MQ (1961) equation is (i) originally derived only for a soil medium with randomly dis tributed particles of uniform size (most resembling a coarse sandy soil), (ii) derived for the case of permeability rather than diffusivity, (iii) not taking into account any effects of soil pore size distribution, and (iv) not validated against undisturbed soil data representing a broad range of soil texture, the almost universal use of the MQ (1961) equation to predict $D_p(\varepsilon)$ does not seem warranted.

Air permeability (k_a) as a function of air-filled porosity (ε) governs convective (pressure gradient induced) air and gas transport in soil. The increased use of soil venting (soil vapor extraction, SVE) systems during vadose zone remediation at polluted soil sites has created a renewed interest in k_a and its dependency on soil type and ε, since k_a typically will be the governing parameter for SVE system performance and clean-up efficiency (Poulsen *et al.,* 1998). Also, air permeability is an easily and rapidly measured parameter compared to for example gas diffusivity (Iversen *et al.,* 2001), and k_a alone or in combination with D_p provides valuable information about soil structure and pore connectivity (Ball, 1981, Moldrup *et al.,* 2001, 2003 a). At present, however, no reliable models to predict k_a as a function of ε in undisturbed soil across soil types are available (Moldrup *et al.,* 1998, 2001, 2003 a).

The objective of this paper is to review, connect, and put into perspective recent tests of models for soil water retention (pore size distribution), gas diffusivity, and air permeability, as compared with data for undisturbed volcanic ash soils (Andisols) from three prefectures in Japan. Further details about data and model tests can be found in Moldrup *et al.* (2003 a, b). The use of combined water retention, gas diffusivity, and air permeability data to provide valuable information about soil aeration, pore characteristics, and soil structure is briefly discussed.

2. Materials and Methods

2. 1 Materials

The 18 Andisols considered here represent different location, soil type, soil depth, and soil management and cultivation, and can briefly be described as follows :

1) Seven Andisols from Tsumagoi, Gunma Prefecture, Honshu (labelled Tsumagoi 1–7). The sample area is characterized by humic and fine-textured Andisols with typically 30–50% clay. The main crop was cabbage (Tsumagoi 1–5). Tsumagoi 6–7 were sampled at a non-cultivated field.

2) Five Andisols from Miura, Kanagawa Prefecture, Honshu (Miura 1–5). The sample area is characterized by light-clay Andisols. The main crop was Japanese radish. Miura 4–5 were sampled at a field where soil layer exchange treatment had taken place 3–4 years previous to sampling.

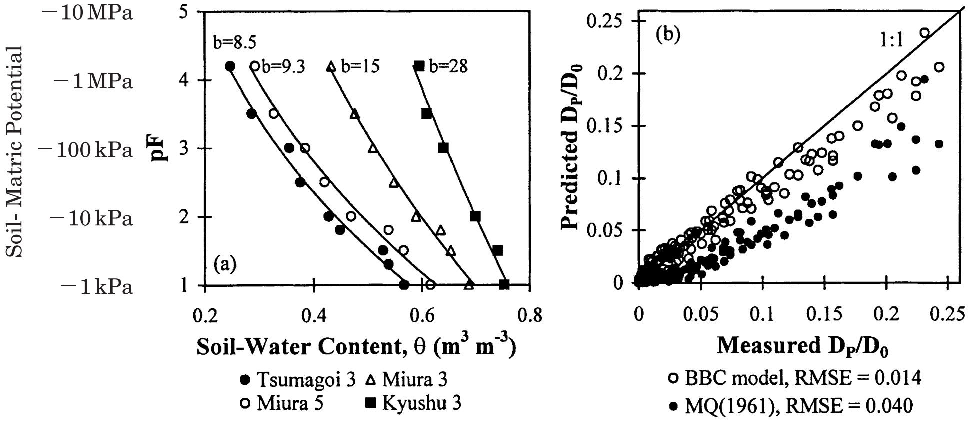

Fig. 1 (a) The Campbell water retention model (Eq. [1]) fitted to measured data for 4 Andisols. pF $= \mathrm{Log}(-\psi_{\mathrm{cm\,H_2O}}) = \mathrm{Log}(-\psi_{\mathrm{Pa}}) - 2$. (b) Scatterplot comparison of predicted and measured relative gas diffusivities (D_p/D_0) for 18 Andisols (Moldrup *et al*, 2003 b). Test of the Millington and Quirk (1961) model (Eq. [2] ; solid circles), and the BBC model (Eq. [3] ; open triangles). RMSE is root mean square error of prediction.

3) Six Andisols from Kumamoto prefecture in Kyushu (Kyushu 1–6). The sample area (grasslands) were characterized by humic (Kyushu 4–6) and highly humic (Kyushu 1–3) Andisols.

2. 2 Measurements

Soil water retention (pore size distribution), gas diffusivity, and air permeability data were measured at between 6 and 9 different soil matric potentials. The measurements were done on undisturbed 100 cm^3 soil samples in a broad matric potential interval from −1 kPa (near water saturation) to −1.5 MPa (wilting point). Three closely-sampled soil cores were used for each soil. Since variability between samples were low, especially for soil water retention and gas diffusivity, mean values for three samples were used in the predictive model tests. Soil water retention was measured following Klute (1986). Gas diffusivity was measured as described by Osozawa (1987), following the principles of Currie (1960). Air permeability was measured following the principles of Grover (1956).

3. Modelling Approaches and Results

3. 1 Soil-Water Retention

We applied the Campbell (1974) soil water retention (pore size distribution) model,

$$\psi/\psi_e = (\theta/\theta_s)^{-b}, \qquad [1]$$

where ψ is soil matric potential (Pa), ψ_e is soil matric potential at air entry, θ is volumetric soil water content, θ_s is volumetric soil water content at water saturation, and b (>0) is the Campbell pore size distribution parameter, corresponding to the slope of the soil water retention curve in a $\mathrm{Log}(\theta)$-$\mathrm{Log}(-\psi)$ coordinate system.

The Campbell model (Eq. [1]) provided a near-perfect fit to the retention data for all 18 Andisols (coefficient of regression, $r^2 > 0.99$) from −1 kPa to −1.5 MPa (Moldrup *et al.,* 2003 b). Values of Campbell b ranged from 8.3 (Miura 2) to 40.8 (Kyushu 2). Examples of the fit by the Campbell (1974) retention model to measured data are shown in Fig. 1 a.

Because of the excellent fit by the simple Campell (1974) model, more complicated (multi-parameter) retention models like the Mualem-van Genuchten (van Genuchten, 1980) model were not considered and, consequently, predictive gas diffusivity models linked to the Mualem-van Genuchten retention model (e.g. : Freijer, 1994) were also not considered. However, we recognize that bimodal water retention models based on the Mualem-van Genuchten or similar unimodal retention models can provide even more accurate fit to soil-water retention data for Andisols, from water saturation to wilting point (Abenney-Mickson *et al.,* 1996). Bimodal water retention models may therefore be highly useful to derive further information on pore size distribution and pore characteristics and to predict unsaturated hydraulic conductivity and water and solute transport in Andisols.

3. 2 Gas Diffusivity

We tested the Millington and Quirk (1961) model,

$$D_p/D_0 = \varepsilon^{3.33}/\Phi^2, \quad [2]$$

where D_p is the gas diffusion coefficient in soil, D_0 is the gas diffusion coefficient in air, ε is the soil air-filled porosity (volumetric soil air content), and Φ is the soil total porosity. Furthermore, we tested two recent and soil-type (Campbell *b*) dependent gas diffusivity models for undisturbed soil. The first is the BBC [Buckingham(1904)-Burdine(1953)-Campbell (1974)] model suggested by Moldrup *et al.* (1999),

$$D_p/D_0 = \Phi^2(\varepsilon/\Phi)^{2+(3/b)}, \quad [3]$$

and the second is the soil macro-porosity dependent model by Moldrup *et al.* (2000),

$$D_p/D_0 = [2(\varepsilon_{-10\,\mathrm{kPa}}{}^3 + 0.04\varepsilon_{-10\,\mathrm{kPa}})] \cdot (\varepsilon/\varepsilon_{-10\,\mathrm{kPa}})^{2+(3/b)}, \quad [4]$$

where $\varepsilon_{-10\,\mathrm{kPa}}$ is the soil air-filled porosity at -10 kPa of soil matric potential, corresponding to the volumetric content of soil pores with an equivalent diameter $>30\mu$m, and is labeled the soil macro-porosity. Moldrup *et al.* (2000) found that the first term in Eq. [4] (marked by brackets) well described gas diffusivity at -10 kPa for 144 undisturbed soil from Europe, and that the entire equation well predicted D_p as a function of ε for further 21 undisturbed European soils where complete $D_p(\varepsilon)$ data sets were available. The BBC model (Eq. [3]) gave equally accurate predictions as Eq. [4], while the MQ (1961) model (Eq. [2]) gave poor predictions except for sandy soils.

When tested against the $D_p(\varepsilon)$ data for the 18 Andisols, the two Campbell *b* dependent models gave similar and good predictions, while the MQ (1961) model markedly under-estimated the measured $D_p(\varepsilon)$ data for all 18 Andisols (Moldrup *et al.,* 2003 b). Figure 1 b shows a comparison between the performance of the BBC model, Eq. [3], and the MQ (1961) model, Eq. [2], tested against the data for the 18 Andisols. The test results emphasize that the soil type independent Millington and Quirk (1961) $D_p(\varepsilon)$ model cannot provide realistic predictions of gas diffusivity in Andisols. Instead, any of the two soil type dependent (Campbell b dependent) $D_p(\varepsilon)$ models can be used to obtain realistic predictions for diffusive gas transport in undisturbed Andisols.

3. 3 Air Permeability

We applied a general power function model for $k_a(\varepsilon)$ with reference point not at air saturation but instead at -10 kPa in order to make the $k_a(\varepsilon)$ model analogous to the above presented gas diffusivity models. Thus,

$$k_a = k_{a,\,-10\,\mathrm{kPa}}(\varepsilon/\varepsilon_{-10\,\mathrm{kPa}})^{\eta}, \quad [5]$$

where η is a tortuosity/connectivity parameter. Based on Moldrup *et al.* (1998, 2001), we tested Eq. [5] with (i) $\eta = 1 + (b/20)$, and (ii) $\eta = 1 + (b/4)$. The original model by Moldrup *et al.* (1998) with $\eta = 1 + (b/4)$ did a poor job in predicting $k_a(\varepsilon)$ for the Japanese Andisols, both in more dry soil (matric potential below -10 kPa) and in more wet soil (matric potential above -10 kPa). This is in agreement with the previous model tests against European soils, where Eq. [5] with $\eta = 1 + (b/4)$ only could predict $k_a(\varepsilon)$ well for sandy soils with b values below 6 (compared to *b* values always exceeding 8 for the Japanese Andisols).

The recent model by Moldrup *et al.* (2001) with $\eta = 1 + (b/20)$ had previously tested well for European soils with clay contents between 11 and 46% and, also, tested well for the Japanese Andisols under wet and medium soil mois-

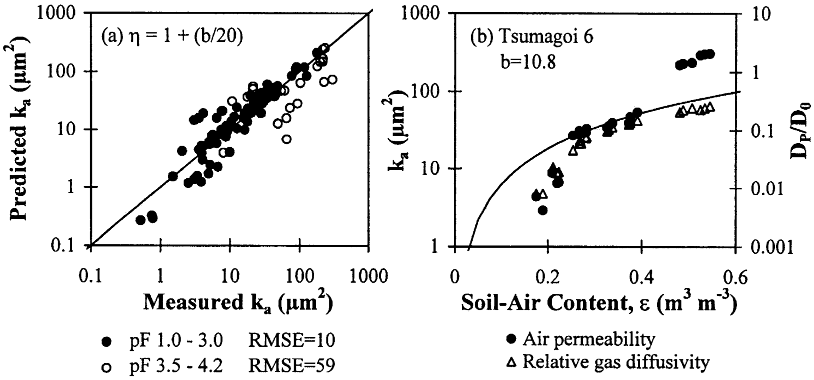

Fig. 2. (a) Scatterplot comparison of predicted (Eq. [5]) and measured air permeabilities for the Andisols (Moldrup *et al.,* 2003 a). (b) Comparison of air permeability (k_a) and gas diffusivity (D_p/D_0) for Tsumagoi 6. The solid line is the predictive $k_a(\varepsilon)$ model (Eq. [5] with $\eta = 1 + (b/20)$).

ture conditions (between −1 kPa and −100 kPa) (Moldrup *et al.,* 2003a). However, for the highly-structured Tsumagoi soils, k_a steeply increased with increasing ε under dry soil conditions, suggesting the sudden occurrence of a highly connected air-filled pore network under drainage. Because of this, the $k_a(\varepsilon)$ model with $\eta = 1 + (b/20)$ markedly under-predicted the measured $k_a(\varepsilon)$ for $\psi < -300$ kPa (pF > 3.5) for some soils, see Fig. 2 a.

The sudden increase in k_a at dry conditions is shown for Tsumagoi 6 in Fig. 2 b. Since the sudden increase in the gas transport parameter and the subsequent under-prediction by a simple power function model is not observed for gas diffusivity (example given in Fig. 2 b), the results clearly imply that k_a is markedly influenced by soil structure while D_p is much less affected. The solid line in Fig. 2 b is the predictive $k_a(\varepsilon)$ model (Eq. [5] with $\eta = 1 + (b/20)$). Note that in Moldrup *et al.* (2003 a), a different (soil type independent) $k_a(\varepsilon)$ model is compared with the same data.

To describe $k_a(\varepsilon)$ across the entire matric potential range, a two- or three-region $k_a(\varepsilon)$ model would be needed to better describe k_a in the dry region (to include soil structure, especially connectivity, effects) and in the wet region (to include effects of pore blocking by interconnected water films), cf. Fig. 2 b. Recently, a two-region probability-law model for $k_a(\varepsilon)$ that can accurately describe $k_a(\varepsilon)$ data for well-structured Andisols (e.g., the Tsumagoi Andisol shown in Fig. 2 b) within a broad matric potential range has been developed (Poulsen *et al.,* 2003).

At present, measurements of k_a at at least one soil matric potential (suggested to be −10 kPa) or preferably more are needed to apply the predictive model [Eq. [5] with $\eta = 1 + (b/20)$] in the wet to medium soil moisture range. Without any k_a measurements, $k_a(\varepsilon)$ can at present not be realistically predicted in undisturbed soils including Andisols.

3. 4 Gaseous Phase Fingerprints

The data for the Japanese Andisols, measured by Yoshikawa, are unique because soil water retention, soil gas diffusivity, and soil air permeability were measured on the same, undisturbed soil samples at as many as 9 different soil matric potentials. This enabled us to develop and test new soil indexes based on the gas transport parameters (D_p and k_a). In Moldrup *et al.* (2003 a, b), two such indexes are suggested :

1) A soil aeration index based on measurements of D_p and soil water retention on the same undisturbed soil samples at a minimum of 6–7 different soil matric potentials. This

so-called Gas Diffusion Fingerprint (GDF) plot provided the ability to clearly distinguish between soils with low and high aeration potential and, thus, to identify soils with a high possibility of developing plant diseases related to poor soil aeration.

2) A soil structure index based on measurements of k_a, D_p, and soil water retention on the same undisturbed soil samples at a minimum of 6–7 different soil matric potentials. This in combination with the Millington and Quirk (1964) convective-diffusive fluid flow model enabled the development of a simple Soil Structure Fingerprint (SSF) plot that clearly showed the effects of soil management and organic matter content on soil structure.

Such soil gaseous phase fingerprints may be valuable in evaluating soil management and cultivation effects on pore network and soil and plant health. Also, combined measurements of water retention, gas diffusivity, and air permeability may provide new information about inactive pore space for gas transport (Schjonning *et al.,* 2002).

4. Conclusions

1) Soil water retention (pore size distribution) from -1 kPa to -1.5 MPa of soil matric potential was well described by the simple Campbell (1974) retention model for all 18 undisturbed Andisols. Although the use of multi-parameter, bimodal retention models seems promising for describing pore size distribution and hydraulic characteristics for Andisols (e.g. : Abenney-Mickson *et al.,* 1996), multi-parameter retention models are likely not necessary in relation to developing accurate predictive models for gas diffusivity.

2) Gas diffusivity in the same matric potential range (-1 kPa to -1.5 MPa) was accurately predicted by two recent soil type (Campbell *b*) dependent models, while the popular Millington and Quirk (1961) model markedly underpredicted $D_p(\varepsilon)$ for all 18 Andisols (Moldrup *et al.,* 2003 b). The Millington and Quirk (1961) model is not realistic for predicting gas diffusivity in undisturbed Andisols. Instead, the Campbell *b* dependent $D_p(\varepsilon)$ models are recommended for use in future gas transport and fate models for Andisols.

3) Air permeability in wet to medium moist soil (-1 to -100 kPa of matric potential) was well predicted by a simple Campbell *b* dependent model, provided that k_a at -10 kPa of matric potential was measured (Moldrup *et al.,* 2003 a). Since -1 to -100 kPa of matric potential is a realistic range for soil venting systems, the *b* dependent $k_a(\varepsilon)$ model seems useful in simulating and designing soil venting systems. Predictions of $k_a(\varepsilon)$ also in more dry Andisol will require two- or multi-region models for $k_a(\varepsilon)$ together with additional k_a measurements.

4) Soil physics has traditionally been dominated by *soil water physics.* The combined used of pore size distribution, gas diffusivity, and air permeability data can give us new information on soil management and cultivation effects on soil aeration, soil structure and soil pore characteristics and pore networks and can open up for new and exiting findings within the area of *soil air physics.*

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不攪乱火山灰土壌における気体移動パラメータ予測に関する最近の研究進展：相対ガス拡散係数と通気係数のCampbell-bパラメータ依存モデル

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要　旨

本論文は，日本の火山灰土壌（アンディソル）についての気体移動パラメータ予測モデルに関する最近の研究をまとめ，モデル適用の展望を示したものである。日本の3つの県の土性の異なった18の火山灰土壌すべてにおいて，土壌マトリックポテンシャル－1 kPa～－1.5 MPa域の水分特性曲線はCampbell（1974）モデルにより，よく記述することができた。同マトリックポテンシャル域での相対ガス拡散係数は，2つの新しい土壌タイプ依存モデル（Campbell-b依存モデル）により，よく予測できた。2つのモデルは，⑴BBC（Buckingham-Burdine-Campbell）モデルと，⑵マクロポロシティ依存モデル（－10 kPaにおける気相率に依存するモデル）である。一方，広く使われているMillington and Quirk（1961）モデルでは，相対ガス拡散係数は18種類の火山灰土壌すべてで低く見積られた。中湿土壌（マトリックポテンシャル－1～－100 kPa）の通気係数k_aは，実測されたマトリックポテンシャル－10 kPaでのk_aを用いることにより，単純なCampbell-b依存モデルにより，よく予測できた。結論として，Campbell-bに基づいたモデルは，火山灰土壌の間隙特性（土壌水分特性曲線，土壌構造指標）を記述し，気体移動パラメタを予測するのに非常に有効であることが明らかとなった。

キーワード：相対ガス拡散係数，ガス移動，Campbell土壌水分保持モデル，BBCガス拡散モデル，土壌構造指標

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Dr. Moldrup 講演に関する質疑

質問：

同じ土壌の不飽和透水係数を求めるとき二元的な土壌孔隙分布のタイプ，または透水係数分布のタイプが見られるように思えるが，その点についてどう考えるか。

Moldrup（オルボー大）：

同じ土壌で不飽和透水係数を何度か計測したことがあるが，いわれるような関係が孔隙分布特性，または水分特性曲線との間に見られた。しかしながら，通気性の場合は，その変化は水の場合と全く異なり bimodal 曲線を示し，孔隙分布との間に明瞭な関係は認められなかった。

質問：

透水係数との関係はどうか。

Moldrup：

不飽和透水係数の場合には，孔隙分布に基づく 2〜3 領域モデルで非常に良く表すことができ，その傾向は Andisol でも変わりはなかった。Andisol の場合，一つの興味ある点が明らかとなった。それは，水が大きな孔隙を主に流れる飽和透水係数と大きな孔隙のみを空気が流れる pF 2.0 通気性との間に非常によい相関が認められることだ。そこで，飽和透水係数が不明の場合，pF 2.0 通気性から飽和透水係数が推定しうるのではないかと考えている。また，通気性測定の場合，水の移動に比べガス移動は非常に速いため透水性測定より短時間で測定できる，透水性測定の時水の注入時にどうしても試料を撹乱しがちであるが，通気性測定の場合その危険性が少ないなどの利点があり，飽和透水係数の計測に利用できないかと考えている。

質問：

ガス拡散 finger print（GDF）と土壌構造 finger print の考え方をわかりやすく説明してもらいたい。

Moldrup：

我々は幾度か異なる土壌マトリックスポテンシャル下での相対的なガス拡散係数を求めてきた。ここで相対的なガス拡散係数とはあるポテンシャルでのガス拡散係数 D_i を大気中での拡散係数 D_0 で割った値（D_i/D_0）で 0〜1 の値を取る。今，仮に土壌マトリックスポテンシャル−10 cm H_2O (1) と−30 cm H_2O (2) があるとするとき，両ポテンシャルでの air filled pore を e_1, e_2, 相対ガス拡散係数を D_1, D_2 とする。そして，それぞれのポテンシャルでの相対拡散係数の差を air filled pore の差で割った値［$(D_2-D_1)/(e_2-e_1)$］を定義する。この値は，マトリックスポテンシャルが変化する間（この場合は−10 cm H_2O から−30 cm H_2O の間）にどの程度のガス拡散に寄与しうる孔隙が土壌中に空くのかを示している。また，言い換えれば，二つのポテンシャル間で増加した air filled pore がどの程度ガス拡散に寄与しているかを示している。今回の報告で用いた Andisol は飽和に近い状態から（−10 cm H_2O）から圃場用水量まで迅速に水が排除され非常に“良い”土壌であるといえる。“良い”とは air filled pore が迅速に増加し，それに伴う拡散係数の増加も顕著であるということを示す。つまり，Andisol は飽和近くから圃場用水量まで迅速な排水が見られ，すぐに高い拡散係数が得られるという意味で“良い”のである。一方，嬬恋で採取された粘土に富む Andisol は同様に air filled pore が増加したときそれに伴う拡散係数の増加が非常に少なく，これは“良くない”土壌であるといえる。このような“良くない”土壌では作物の根の生育に障害がでることが予測され，実際に作物の根に病気が発生していると聞いている。このように実際の農業の現場において，一般的なモデルを用いて計算するのではなくより簡潔に土壌がガス拡散に有効な air filled pore を生じやすいのかどうか判定しうる考え方が必要であり，これが上で述べたガス拡散 finger print の考え方である。この概念は確かに非常に単純化しすぎていると感じるかも知れない。確かに，これは土壌の孔隙構造として 1964 年に Millinton and Quirk が示した平行管モデルを基にしているに過ぎない。しかし，この指標は土壌の孔隙構造を知るための第 1 次のラフなインデックスと我々は位置づけている。例えば，ある土壌の平均等価径が大きい場合には，そこには連続した大孔隙径が発達しているものと期待することはできる。この指標から必要以上の多くの情報を得ることは期待すべきではないが，少なくともこの指標を通じて，有機物に富むこの Andisol という土壌が，非常に大きな通気性を示しうるものであることは知ることができる。

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