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# Soil dielectric permittivity modelling for 50 MHz instrumentation

Gaston Mendoza Veirana<sup>a,\*</sup>, Jeroen Verhegge<sup>a,b</sup>, Wim Cornelis<sup>a</sup>, Philippe De Smedt<sup>a,b</sup>

<sup>a</sup> Department of Environment, Faculty of Bioscience Engineering, Ghent University, Coupure Links 653, geb. B, 9000 Ghent, Belgium
 <sup>b</sup> Department of Archaeology, Ghent University, Sint-Pietersnieuwstraat 35-UFO, 9000 Ghent, Belgium

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# ABSTRACT

Near surface electromagnetic geophysical techniques are proven tools to support soil ecosystem services and soil exploration. Such geophysical techniques provide electromagnetic properties that are useful to characterize the studied soil. The link between relevant soil characteristics and geophysical properties, such as dielectric permittivity ( $\epsilon$ ), is commonly expressed by pedophysical models. However, some weaknesses remain in their application, such as the requirement of parameters that are difficult to measure or calculate. Therefore, these parameters are frequently fixed, but this oversimplifies the complexity of the investigated soils. Moreover, the validity of  $\epsilon$  pedophysical models in the frequency range of operating soil moisture sensors (normally < 100 MHz) remains poorly investigated.

In this study, the accuracy and adaptability of  $\varepsilon$  pedophysical models at different electromagnetic frequency ranges was tested and improved using newly collected laboratory and field data. Such data was collected on soils over a wide range of textures, physical and chemical properties.

To achieve this, we review the measurement methods and characteristics of  $\varepsilon$  pedophysical models, soil phases and geometric parameters. Subsequently, we show how geometric parameters can explain the dependance of soil texture on  $\varepsilon$  by implementing pedotransfer functions. Then, drawing on a broad experimental basis of common soil types in Europe, we develop novel  $\varepsilon$  pedophysical models at 50 MHz. These models are not only easy to evaluate but also capture most of the soil's complexity. Additionally, these new  $\varepsilon$  pedophysical models eliminate the need for calibration data due to the introduction of novel pedotransfer functions based on soil cation exchange capacity. An extensive model test shows an unprecedented decrease in the RMSE of the newly proposed models of up to 412%.

In conclusion, despite it is unlikely to characterize soil structure, bulk density, or temperature at 50 MHz, these updated PPMs are useful for highly accurate water content and  $\varepsilon$  predictions, in both laboratory and field conditions, without the need for calibration data. As the developed modelling procedures are valid for a wide range of electromagnetic frequencies, these can be applied to soil exploration with TDR and GPR instrumentation.

For reproducibility, all collected soil data are provided, alongside open-source Python code that contains the presented modelling procedures.

# 1. Introduction

The applications of electromagnetic geophysical methods for investigating soils are continually increasing. This is due to their ability to provide quick and high-resolution data of the soil surface. Ground penetrating radar (GRP), electromagnetic induction method (EMI), and resistivity methods surveys are among these techniques. Other methods based on hand-probes instruments such as time domain reflectometry (TDR), time domain transmissions (TDT), and impedance and capacitance sensors, are suitable for precise moisture readings and collecting continuous monitoring data. Such methods inform about soil geophysical properties as bulk real electrical conductivity ( $\sigma_b$ ) and bulk real relative dielectric permittivity ( $\varepsilon_b$ ). The most commonly targeted soil property is the soil water content, for which the measurement of  $\varepsilon_b$  is particularly useful. To meet the growing demand for soil applications, it is necessary to improve and test models that support field investigations for environmental applications. These include contaminant studies (Comegna et al., 2019), soil volumetric water content ( $\theta$ ) estimation

\* Corresponding author. *E-mail address:* gaston.mendozaveirana@ugent.be (G. Mendoza Veirana).

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(Topp et al., 1980; Roth et al., 1990; Wunderlich et al., 2013), soil structure characterization (Romero-Ruiz et al., 2018), and Martian soil characterization (Stillman and Olhoeft, 2008). Owing its widespread application to the strong relationship with (bound and free) soil water (Cassidy and Jol, 2009),  $\varepsilon_b$  is also influenced by soil properties such as soil texture (Bouksila et al., 2008; Jacobsen and Schjønning, 1993; Peplinski et al., 1995), salinity (Seyfried and Murdock, 2004), cation exchange capacity (*CEC*) (Ojo et al., 2015), dry bulk density ( $b_d$ ) (Hollis et al., 2012), porosity ( $\emptyset$ ) pore connectivity (Linde et al., 2006), and particle shape (Cosenza et al., 2009; Sen et al., 1981). Within its complexity,  $\varepsilon_b$  also depends on the frequency of the applied electromagnetic (EM) wave (Jones et al., 2005; Rinaldi and Cuestas, 2002). Combined, these factors pose significant challenges in soil characterization, which can result in imprecise models that may lead to misprediction of soil properties.

Similar to petrophysical models, which link geophysical properties to rock properties, pedophysical models link geophysical to soil properties (and state variables) (Romero-Ruiz et al., 2018). Pedophysical permittivity models (PPMs) are models that link soil properties to  $\varepsilon_b$ . Some of these incorporate the volume proportion of each soil phase (liquid, solid and gas), and their dielectric permittivity (Wunderlich et al., 2013). PPMs also depends on geometrical parameters, which represents the arrangement and shape of soil phases. Importantly, these models are not to be confused with pedotransfer functions (PTFs), which describe relationships between different (sets of) soil properties (Van Looy et al., 2017). We also refer as PTFs the models that link different geometrical parameters.

Despite the wide range of available PPMs in literature, the current state of pedophysical permittivity modelling presents three key challenges. First, available PPMs depend on parameters that are difficult to quantify in the laboratory or field, such as geometric parameters and the permittivity of the soil phases. It is also unclear to what extent these parameters influence  $\varepsilon_b$  at different EM frequencies. Second, these parameters are usually assumed as constants for all samples (fixed), which underestimates the variability of investigated soils, or they are fitted to calibration  $\varepsilon_b$  data (if available), potentially masking errors during the fitting process. Lastly, the applicability of PPMs for the frequency range in which impedance and capacitance moisture sensors operate (20 to 100 MHz) remains poorly investigated.

In this study, we aim to overcome these challenges by first reviewing existing laboratory and modelling methods to obtain relevant soil properties involved on PPMs. Using these methods, we obtained new  $\varepsilon_b$  data and soil properties from a collection of soil samples, reflecting the variability of European soils. The data were collected using an impedance moisture probe in field and laboratory conditions at 50 MHz. These data are then used to evaluate a large list of available PPMs. To shed light on geometrical parameter estimation, a test of available and new PTFs is presented. Thus, through a semi-empirical approach that integrates adaptive PTFs, we develop and test new PPMs using the new  $\varepsilon_b$  data. Finally, to evaluate the influence of each soil variable on  $\varepsilon_b$ , a sensitivity analysis is performed on the most accurate new PPM.

# 2. Pedophysical modelling

# 2.1. Commonly used soil pedophysical dielectric permittivity models

To provide insight into how PPMs are formulated, first, the relative dielectric permittivity of a bulk soil has to be defined. Generally, this is a complex unit  $(\epsilon_b^*)$ :

$$\varepsilon_b^* = \varepsilon_b - j\varepsilon_b^{'} \tag{1}$$

with the imaginary unit *j* and  $\varepsilon_b^{"}$  the soil bulk imaginary relative dielectric permittivity. While most PPMs are expressed in terms of  $\varepsilon_b$ , some use the soil bulk apparent relative dielectric permittivity ( $\varepsilon_{ap}$ ):

$$\varepsilon_{ap} = \frac{\varepsilon_b}{2} \left( 1 + \sqrt{1 + \left(\frac{\varepsilon_b^*}{\varepsilon_b}\right)^2} \right)$$
(2)

Based on the nature of their development, PPMs can be grouped into three classes: empirical, volumetric mixing, and effective medium (Romero-Ruiz et al., 2018; Wunderlich et al., 2013). A first class comprises empirical models, which are generally developed by fitting a polynomial to experimental (laboratory or field) data. Empirical PPMs were often developed for specific soil environments using data obtained with TDR sensors, which normally provides  $\varepsilon_{ap}$  instead of  $\varepsilon_b$ . Examples include the Topp equation (Eq. (3) and the model proposed by Jacobsen and Schjønning (1993) (Eq. (4):

$$\theta = -5.3 \bullet 10^{-2} + 2.92 \bullet 10^{-2} \varepsilon_{ap} - 5.5 \bullet 10^{-4} \varepsilon_{ap}^2 + 4.3 \bullet 10^{-6} \varepsilon_{ap}^3$$
(3)

$$\theta = -3.41 \bullet 10^{-2} + 3.45 \bullet 10^{-2} \varepsilon_{ap} - 11.4 \bullet 10^{-4} \varepsilon_{ap}^{2} + 17.1 \bullet 10^{-6} \varepsilon_{ap}^{3} - 3.7$$
  

$$\bullet 10^{-2} b_{d} + 7.36 \bullet 10^{-4} cc \bullet 100 + 47.7 \bullet 10^{-4} org \bullet 100$$
(4)

where *org* is the soil organic matter content and *cc* ([g/100 g]) the soil clay content. Such models are widely used because of their simplicity, straightforward application in a site-specific context, and through their polynomial coefficients show the relevance of the involved properties. A primary weakness of empirical PPMs is their frequent poor performance beyond the initial empirically tested conditions (Wunderlich et al., 2013).

Secondly, volumetric mixing (or volume averaged) are PPMs where  $\varepsilon_b$  is calculated by combining soil phase components (typically water, solid, and air), their fractional volume, and geometrical arrangement. The most used volumetric mixing PPM was proposed by Lichtenecker and Rother (1931) (LR):

$$\varepsilon_{b} = \left(\theta\varepsilon_{w}^{a} + (1-\emptyset)\varepsilon_{s}^{a} + (\emptyset-\theta)\varepsilon_{a}^{a}\right)^{1/a}$$
(5)

If  $\alpha = 0.5$  in Eq. (5), this becomes:

$$\varepsilon_b = \left(\theta \varepsilon_w^{0.5} + (1 - \emptyset) \varepsilon_s^{0.5} + (\emptyset - \theta) \varepsilon_a^{0.5}\right)^2 \tag{6}$$

where  $\alpha$  is a geometrical parameter,  $\varepsilon_w$  the real relative dielectric permittivity of the water soil phase,  $\varepsilon_s$  the real relative dielectric permittivity of the solid soil phase, and  $\varepsilon_a$  the real relative dielectric permittivity of the air soil phase. When Eq. (5) is replaced by  $\alpha = 0.5$  this becomes the complex refractive index model (CRIM) (Eq. (6) (Birchak et al., 1974; Roth et al., 1990). Another relevant volumetric mixing PPM was proposed by Linde et al. (2006):

$$\varepsilon_b = \mathcal{Q}^m \left( S^n \varepsilon_w + (\mathcal{Q}^{-m} - 1) \varepsilon_s + (1 - S^n) \varepsilon_a \right)$$
(7)

If m = 1.5 and n = 2 in Eq. (7), this becomes:

$$\varepsilon_b = \emptyset^{1.5} \left( S^2 \varepsilon_w + \left( \emptyset^{-1.5} - 1 \right) \varepsilon_s + (1 - S^2) \varepsilon_a \right)$$
(8)

where  $S (=\theta/\emptyset)$  is the soil saturation, and the geometrical parameters *m* (cementation factor) and *n* (saturation factor) are similar to those proposed by Archie (1942). Empirical estimations of *m* and *n* found normal values of 1.5 and 2 (Romero-Ruiz et al., 2018; Glover, 2015), respectively, composing the Eq. (8).

The main advantage of volumetric mixing PPMs is the deeper insight into the  $\varepsilon_b$  estimation by accounting for phases arrangement. Also, volumetric mixing models allow a more straightforward integration of soil characteristics because they generally involve a larger set of soil properties and model parameters than their empirical counterparts. Nevertheless, this flexibility constitutes also the main downside of volumetric mixing models, because their application is challenging in soils with some unknown properties (e.g., soil porosity).

Lastly, effective medium models, also known as embedding schemes, are physically-based schemes whereby one or more soil phases are progressively embedded into the bulk soil being represented by a homogeneous background material (e.g., Cosenza et al., 2009; Sen et al., 1981; Wunderlich et al., 2013). The embedding process is simulated by differential steps that ranges from initial to actual conditions. An example of homogeneous dry soil background with embedding water particles is shown in Wunderlich et al. (2013):

$$\frac{d\varepsilon(p)}{dp} = \frac{\varepsilon(p)(\theta_n - \theta_1)}{1 + \theta_1 - \theta_n + p(\theta_n - \theta_1)} \frac{\varepsilon_w - \varepsilon(p)}{L_w \varepsilon_w + (1 - L_w)\varepsilon(p)}$$
$$\varepsilon(p = 0) = \varepsilon_1; \varepsilon(p = 1) = \varepsilon_b$$
(9)  
If  $L_w = 0.01$  in Eq. (9), this becomes:

$$\frac{d\varepsilon(p)}{dp} = \frac{\varepsilon(p)(\theta_n - \theta_1)}{1 + \theta_1 - \theta_n + p(\theta_n - \theta_1)} \frac{\varepsilon_w - \varepsilon(p)}{0.01\varepsilon_w + 0.99\varepsilon(p)}$$
$$\varepsilon(p = 0) = \varepsilon_1; \varepsilon(p = 1) = \varepsilon_b \tag{10}$$

where the geometrical parameter  $L_w$  is the depolarization factor of the soil water phase (normally fixed as 0.01, Wunderlich et al. (2013)). p is the integration variable that ranges from 0 to 1,  $\varepsilon(p = 0) = \varepsilon_1$  is the initial  $\varepsilon_b$  that corresponds to the initial volumetric water content ( $\theta_1$ ); similarly,  $\varepsilon(p = 1) = \varepsilon_b$  corresponds to  $\theta$ . A last example is the PPM shown by Endres and Redman (1996), where firstly air is embedded into water, after which solid grains are added to this air–water background:

$$\frac{d\varepsilon(p)}{dp} = \frac{\varepsilon(p)(1-S)}{S+p(1-S)} \frac{\varepsilon_a - \varepsilon(p)}{L\varepsilon_a + (1-L)\varepsilon(p)}$$

$$\varepsilon(p=0) = \varepsilon_w; \ \varepsilon(p=1) = \varepsilon_p$$

$$\frac{d\varepsilon(p)}{dp} = \frac{\varepsilon(p)(1-\emptyset)}{\emptyset + p(1-\emptyset)} \frac{\varepsilon_s - \varepsilon(p)}{L\varepsilon_s + (1-L)\varepsilon(p)}$$

$$\varepsilon(p=0) = \varepsilon_p; \ \varepsilon(p=1) = \varepsilon_b$$
If  $L = 0.3$  in Eq. (11), this becomes:
$$(11)$$

$$\frac{d\varepsilon(p)}{dp} = \frac{\varepsilon(p)(1-S)}{S+p(1-S)} \frac{\varepsilon_a - \varepsilon(p)}{0.3\varepsilon_a + 0.7\varepsilon(p)}$$
$$\varepsilon(p=0) = \varepsilon_w; \ \varepsilon(p=1) = \varepsilon_p$$
$$\frac{d\varepsilon(p)}{dp} = \frac{\varepsilon(p)(1-\emptyset)}{\emptyset + p(1-\emptyset)} \frac{\varepsilon_s - \varepsilon(p)}{0.3\varepsilon_s + 0.7\varepsilon(p)}$$
$$\varepsilon(p=0) = \varepsilon_p; \ \varepsilon(p=1) = \varepsilon_b$$
(12)

where the geometrical parameter *L* is the depolarization factor of solid particles,  $\varepsilon_p$  is the real bulk relative dielectric permittivity of the air—water background. Embedding schemes are particularly accurate if some of the incorporated variables, such as the depolarization factor, are fitted to calibration data. Additional PPMs are listed in Table A1.

Despite the complexity of the presented PPMs, these do not explicitly consider the EM frequency dependence of  $\varepsilon_b$ , or soil texture. This can lead to important estimation errors (e.g., misestimating  $\theta$  in clayey soils by 25%) (Le, 2018; Ojo et al., 2015). Accounting for these two variables is therefore crucial to develop well-suited and accurate PPMs.

## 2.2. Soil texture influence in dielectric dispersion

The frequency dependence of  $\varepsilon_b$  (dielectric dispersion) is complex because of the different dielectric responses of the soil phases and their interaction (González-Teruel et al., 2020). Particularly, a dielectric loss mechanism is of especial interest, the Maxwell-Wagner (M-W) effect (or interfacial polarization). The extent of the M-W effect on  $\varepsilon_b$  is strongly dependent on  $\sigma_b$ . In the EM spectrum of a soil sample, the relationship between  $\varepsilon_b$  and  $\sigma_b$  is directly proportional below a specific crossover (or transition) frequency ( $f_{co}$ ). However, above  $f_{co}$ , the relationship becomes inversely proportional and more pronounced. In general, the higher the clay content, *CEC*, temperature (*T*), and salinity, the higher  $\sigma_b$  (Glover, 2015). Fig. 1 illustrates the impact of certain soil properties, such as texture (Fig. 1A) and temperature (Fig. 1B), on  $\varepsilon_b$  influenced by  $\sigma_b$ . For different soils,  $f_{co}$  is commonly found to be between 100 and 200 MHz (Cassidy and Jol, 2009; Chen and Or, 2006a; Jones et al., 2005).

In soil dielectric permittivity applications, TDR, TDT, and GPR typically operate at frequencies above  $f_{co}$ . While the frequency range below  $f_{co}$  includes most impedance and capacitance-based dielectric sensors, which are generally cheaper and can be used to explore all sorts of soils (Vaz et al., 2013). Unlike GPR applications, that are more expensive and are limited in highly conductive soils due to attenuation effects.

In summary, it is expected that clayey samples have higher  $\varepsilon_b$  at frequencies below  $f_{co}$  (Ojo et al., 2015), while sandy samples have higher



Fig. 1. Frequency dependence of  $\varepsilon_b$  with varying  $\sigma_b$ . In Fig. 1A, clay (sodium bentonite), sand (quartz), and talc samples were analyzed using a network vector analyzer (adapted from Jones et al., 2005, with permission). The graph shows that a transition occurs around 100 MHz. Below this frequency, the wet clay sample has a higher  $\varepsilon_b$  compared to the wet sand sample, while the opposite is observed above this frequency. Fig. 1B demonstrates how changing  $\sigma_b$  through temperature affects  $\varepsilon_b$  on a sandy loam sample (with aporosity of 0.44) saturated with a 1.551 S/m solution. At frequencies above  $f_{co}$ ,  $\sigma_b$  is proportional to  $\varepsilon_b$ , while an inverse and more pronounced proportion is observed at frequencies below  $f_{co}$ . The graph considers the correction for electrode polarization (adapted from Chen and Or, 2006b, with permission). The frequency at which the relationship between  $\varepsilon_b$  and  $\sigma_b$  switches (i.e., crossover frequency,  $f_{co}$ ) is clearly visible in the graph.

 $\varepsilon_b$  at frequencies above  $f_{co}$  (Hallikainen et al., 1985).

### 2.3. Soil parameters

Soil parameters involved in most PPMs, such as the volumetric fraction of soil phases, their arrangement and dielectric permittivity, can be quantified with different methods. However, to avoid inconsistent use of resultant values and disagreement on their underlying meaning, these parameters have to be clearly defined. Furthermore, their influence on  $\varepsilon_b$  and their frequency dependence also have to be taken into account.

#### 2.3.1. Free and bound water phase permittivity

The soil volumetric water content can be dived depending on the energy status. The volumetric bound water content ( $\theta_{bw}$ ), often considered as a fourth soil phase in PPMs (Dobson et al., 1985), quantifies the water which is rotationally hindered by adsorption to clay particles surface.

The real relative dielectric permittivity of bound water ( $\varepsilon_{bw}$ ) is significantly lower than that of free water (i.e.,  $\varepsilon_w$ ) due to the molecular behavior of water, which possesses a dipole moment, in the presence of an EM field (Kameyama and Miyamoto, 2008). This behavior, and thus the resulting permittivity, depends on the water's temperature, salinity, and EM frequency. Despite the influence of these factors, most PPMs applications employ a fixed  $\varepsilon_w$  of 80 or 81 (e.g., Kelleners and Verma, 2010; Steelman and Endres, 2011; Wunderlich et al., 2013).

To account for temperature influences on  $\varepsilon_w$ , empirical formulas such as that of Malmberg and Maryott (1956), are used at frequencies below 100 MHz:

$$\varepsilon_w(T) = a_0 + a_1 T + a_2 T^2 + a_3 T^3 \tag{13}$$

With the empirical parameters  $a_0=87.740$ ,  $a_1=-0.40008$  °C<sup>-1</sup>,  $a_2=9.398*10^{-4}$  °C<sup>-2</sup>,  $a_3=-1.410*10^{-6}$  °C<sup>-3</sup>, and *T* is in °C.

For frequencies below  $f_{co}$ , the empirical equation provided by Jones et al. (2005) is used occasionally (e.g., Stevens Water Monitoring systems, 2008), even it is not specified the frequency range for which it was developed.

Although Or and Wraith (1999) proposed a detailed semi-empirical method to calculate  $\varepsilon_{bw}$ , this demands knowledge of soil-specific surface area (SSA) and mineralogy. Hence, several researchers have assigned fixed  $\varepsilon_{bw}$  values, specifically 10 (Cosenza and Tabbagh, 2004), 3.2 (similar to ice, Kameyama and Miyamoto, 2008), and 35 (Dobson et al., 1985). Others derived  $\varepsilon_{bw}$  using polynomial curve-fitting (Young et al., 1997; Wunderlich et al., 2013), yielding values from 2.78 to 69, respectively. With the absence of SSA and mineralogy data, these methodologies lead to significantly different  $\varepsilon_{bw}$  values, limiting PPM applications.

#### 2.3.2. Solid phase permittivity

The permittivity of the soil solid phase ( $\varepsilon_s$ ) depends on the soil's texture, organic matter (Cassidy and Jol, 2009), mineralogy (Glover, 2015) and EM frequency, while temperature only has a negligible effect (Seyfried and Murdock, 2004).

Various studies using PPMs typically fix  $\varepsilon_s$  at 5 (e.g., Kelleners and Verma, 2010; Steelman and Endres, 2011; Wunderlich et al., 2013, who follow Cassidy and Jol, 2009). Other authors have opted to infer  $\varepsilon_s$  through a fitting process of  $\varepsilon_b$  (Leão et al., 2015; Young et al., 1997) or by deriving it from the particle density (Dobson et al., 1985).

Despite the common use of a single fixed value,  $\varepsilon_s$  does influence the PPM's performance. For example, varying  $\varepsilon_s$  between 4 and 6 when predicting  $\varepsilon_b$  with the CRIM model leads to a misestimation of 2% (Steelman and Endres, 2011). Moreover, measure  $\varepsilon_s$  is an indirect frequency-dependent process and different protocols have shown similar values.

Determining  $\varepsilon_s$  in laboratory is usually done using the 'two-phase'

method, which consists of measuring  $\varepsilon_b$  in dry soil, and  $\varepsilon_s$  is calculated by solving a volumetric mixing model. While its dependence on a given pedophysical model can introduce additional uncertainty (Robinson and Friedman, 2003), the two-phase method is equally accurate to the immersion method (Robinson and Friedman, 2003) if used with the CRIM model for dry conditions (Eq. (14) (Kameyama and Miyamoto, 2008).

$$\varepsilon_s = \left(\frac{\varepsilon_b^{0.5} - \varepsilon_a^{0.5} \varnothing}{1 - \varnothing}\right)^2 \tag{14}$$

Because this method focuses solely on two phases (solid and air), any potential third phases such as  $\theta_{bw}$ , should be eliminated from the measurements to ensure accuracy (Kameyama and Miyamoto, 2008).

# 2.3.3. Porosity and particle density

All PPM depends on  $\emptyset$ , which is calculated using the dry bulk density  $(b_d)$  (i.e., mass of the bulk soil per unit volume after oven-drying) and the particle density  $(p_d)$  (i.e., the mass of the solid fraction over its volume) through:

$$\emptyset = 1 - \frac{b_d}{p_d} \tag{15}$$

For pedophysical model application,  $p_d$  is normally fixed at 2.65 g/  $cm^3$ . This avoids time-consuming laboratory determination of  $p_d$  with a pycnometer (Blake and Hartge, 1986), that have shown ranges between 2.53 g/ $cm^3$  in sand and 2.86 g/ $cm^3$  in clay (Schjønning et al., 2017). Estimation of  $p_d$  are facilitate by using PTFs that involve soil properties as sand, clay and organic matter content. The most accurate  $p_d$  PTF is that proposed by Schjønning et al. (2017), which has a negligible error for PPM evaluation (i.e., a RMSE of 0.011 g/ $cm^3$ ).

#### 2.4. Interpreting geometric parameters

Geometric parameters of PPMs represent the soil geometrical arrangement and shape of soil phases. Though they highly affect the outcome of the PPM, they are normally oversimplified by using fixed values.

The volumetric mixing PPM proposed by Linde et al., (2006) (Eq. (7) depends on the geometrical parameters *m* and *n*. These were originally defined in Archie's first and second law (Archie, 1942). Despite that *m* and *n* are typically fixed as 1.5 and 2, respectively, these are frequency dependent (Glover, 2015), and their physical meaning is exclusively geometrical (Brovelli and Cassiani, 2008; Glover, 2009; Feng and Sen, 1985). As Eq. (7) suggests, the sensitivity of  $\varepsilon_b$  to porosity increases with increasing *m*, and is associated with higher values of tortuosity (lower connectivity) (Glover, 2009).

Empirical PTFs for *m* were developed using the *CEC* of soils (Mohamed and Paleologos, 2017), and for *n* using the soil clay content (McBratney et al., 2005). Rinaldi and Cuestas (2002) concluded that for silty loam samples with  $b_d > 1.52$  g/cm<sup>3</sup>, *n* approaches *m*. Shah and Singh (2005) considered this similarity of *m* and *n* when developing an empirical PTF for *m* using clay content, based on 30 soil samples (Fig. 2A):

$$m = 0.92(100cc)^{0.2}$$
 for  $cc > 0.05$ ;  $m = 1.25$  for  $cc < 0.05$ ;  $m = n$  (16)

The geometric parameter  $\alpha$ , depends on the orientation of the soil phases from perpendicular ( $\alpha = -1$ ) to parallel ( $\alpha = 1$ ) with respect to the applied EM field (Roth et al., 1990). Dobson et al. (1985) obtained  $\alpha = 0.65$  by regression from data measured at 4 to 18 GHz and soil types ranging from sandy loam to silty clay. Roth et al. (1990) derived  $\alpha = 0.46$  using several soils, and suggest to use a fixed  $\alpha = 0.5$ . While fixed  $\alpha$  values can be representative for certain textures and frequencies, Ponizovsky et al. (1999) demonstrated using TDR that  $\alpha$  was correlated to clay content. This was empirically confirmed by Wunderlich et al. (2013) (Fig. 2B), who obtained  $\alpha$  by fitting  $\varepsilon_b$  data measured with GPR at 1.6 GHz:



adapted from Guillemoteau et al. (2012)



 $\alpha = -0.46cc + 0.71$ 

(17)

Understanding the link between *m* and  $\alpha$  is relevant in cases where  $\sigma_b$  and  $\varepsilon_b$  are modelled in the same soil. Pedotransfer functions linking *m* and  $\alpha$  were widely discussed (see e.g., Romero-Ruiz et al., 2018; Glover, 2009; Brovelli and Cassiani, 2008). In saturated conditions, Brovelli and Cassiani (2008) express it as:

$$m = 1/\alpha \tag{18}$$

These authors concluded that  $\alpha$  is influenced by the ratio between  $\varepsilon_s$  and  $\varepsilon_w$ , and consequently also m. Equation (18) implicitly assumes that  $\frac{\sigma_b}{\sigma_w} = \frac{\varepsilon_b}{\varepsilon_w}$ , and explicitly states  $\frac{\varepsilon_s}{\varepsilon_w} = 0$ , as the observed ratio was considered negligible (i.e., 0.05 (Brovelli and Cassiani, 2008)). However, common moisture sensors (e.g., Stevens Water Monitoring systems, 2008) often use:

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$$\frac{\sigma_b}{\sigma_w} = \frac{\varepsilon_b - \varepsilon_{offset}}{\varepsilon_w}$$
(19)

(Hilhorst, 2000), where  $\sigma_b$  and  $\varepsilon_b$  are measured at the same frequency, and  $\varepsilon_{offset}$  is defined as the intercept of a fitted line between  $\sigma_b$  and  $\varepsilon_b$  (commonly fixed between 2 to 4.5). Similarly, for saturated conditions, by considering the  $\varepsilon_{offset}$  and  $\frac{\varepsilon_s}{\varepsilon_w}$ , *m* can be expressed as:

$$m = \frac{\log\left[\left((1-\emptyset)\left(\frac{\varepsilon_s}{\varepsilon_w}\right)^a + \emptyset\right)^{\frac{1}{a}} - \frac{\varepsilon_{offset}}{\varepsilon_w}\right]}{\log\left[\emptyset\right]}$$
(20)

Thus, Eq. (18) can be considered as a particular case of Eq. (20). Another expression of *m* in function of clay is obtained by combining Eq. (20) and (17):

$$m = \frac{\log\left[\left((1-\emptyset)\binom{\underline{e_{i}}}{\underline{e_{w}}}\right)^{-0.46cc+0.71} + \emptyset\right)^{\frac{1}{-0.46cc+0.71}} - \frac{\underline{e_{offwr}}}{\underline{e_{w}}}\right]}{\log\left[\emptyset\right]}$$
(21)

And by combining Eq. (18) and (17):

$$m = \frac{1}{-0.46cc + 0.71} \tag{22}$$

Although the three PTFs plotted in Fig. 2A are semiempirically formulated based on different data sets, they result in similar trends between m and clay content.

Generally, the depolarization factor L used in the effective medium

$$L = \frac{-1}{m} + 1 \tag{23}$$

Or by combining Eq. (23) with Eq. (18):

$$L = -\alpha + 1 \tag{24}$$

Fig. 2C shows in accordance PTFs for *L* in function of the clay content. The common trend corroborates the intuition that the average solid particle shape tends to flatten with increasing clay content. It should be noted that, as Eq. (16) assumes m = n, the shape of both solid and liquid inclusions is considered equal in effective medium theory (Feng and Sen, 1985).

# 2.5. New PPMs: Accounting for frequency range and soil texture without calibration data

In the last section, existent and newly proposed PTFs show a link between geometrical parameters and soil texture (particularly, clay content) for frequencies above the  $f_{co}$ . These PTFs are then combined with PPMs:

If in Eq. (5) (LR PPM),  $\alpha$  is calculated by Eq. (17), this becomes:

$$\varepsilon_{b} = \left(\theta\varepsilon_{w}^{-0.46cc+0.71} + (1-\emptyset)\varepsilon_{s}^{-0.46cc+0.71} + (\emptyset-\theta)\varepsilon_{a}^{-0.46cc+0.71}\right)^{1/(-0.46cc+0.71)}$$
(25)

Similarly, if in Eq. (7) (Linde PPM), m and n are similar and calculated by Eq. (21), this becomes:

$$\varepsilon_{b} = \theta^{\frac{\log\left[\left(\left(1-\phi\right)\left(\frac{\varepsilon_{s}}{F_{W}}\right)^{-0.46cc+0.71}+\phi\right)^{\frac{1}{-0.46cc+0.71}}-\frac{\varepsilon_{offset}}{F_{W}}\right]}{\log[\phi]}}(\varepsilon_{w}-\varepsilon_{a}) + \phi^{\frac{\log\left[\left(\left(1-\phi\right)\left(\frac{\varepsilon_{s}}{F_{W}}\right)^{-0.46cc+0.71}+\phi\right)^{\frac{1}{-0.46cc+0.71}}-\frac{\varepsilon_{offset}}{F_{W}}\right]}{\log[\phi]}}(\varepsilon_{a}-\varepsilon_{s}) + \varepsilon_{s}$$

$$(26)$$

theory is a three-dimensional vector that characterizes the shape of the embedded solid particles, independently of their size. For isotropic particles, L is a scalar between 0 and 1 related to the axis perpendicular to the applied EM field. Effective medium theory PPMs consider the

Lastly, if in Eq. (11) (Endres-Redman PPM), *L* is calculated by Eq. (23) and Eq. (21), this becomes:

$$\frac{d\varepsilon(p)}{dp} = \frac{\varepsilon(p)(1-S)}{S+p(1-S)} \frac{\varepsilon_a - \varepsilon(p)}{\left(\frac{-\log[\emptyset]}{\log\left[\left((1-\emptyset)\left(\frac{\varepsilon_s}{\varepsilon_w}\right)^{-0.46cc+0.71} + \emptyset\right)^{\frac{1}{-0.46cc+0.71}} + \emptyset\right)^{\frac{1}{-0.46cc+0.71}} + 1\right)}\varepsilon_a + \left(\frac{\log[\emptyset]}{\log\left[\left((1-\emptyset)\left(\frac{\varepsilon_s}{\varepsilon_w}\right)^{-0.46cc+0.71} + \emptyset\right)^{\frac{1}{-0.46cc+0.71}} - \frac{\varepsilon_{afber}}{\varepsilon_w}\right]}\right)\varepsilon(p)$$

macroscopic anisotropy of the soil by the depolarization factor. Since L represents the average shape of embedded solid particles (or water particles,  $L_w$ ), it is difficult to determine it experimentally in the laboratory. Consequently, this parameter is usually determined by curve-

$$\varepsilon(p=0) = \varepsilon_w; \varepsilon(p=1) = \varepsilon_p$$

$$\frac{d\varepsilon(p)}{dp} = \frac{\varepsilon(p)(1-\emptyset)}{\emptyset+p(1-\emptyset)} \frac{\varepsilon_s - \varepsilon(p)}{\left(\frac{-\log[\emptyset]}{\log\left[\left((1-\emptyset)\left(\frac{\varepsilon_s}{\varepsilon_w}\right)^{-0.46cc+0.71} + \emptyset\right)^{-0.46cc+0.71} - \frac{\varepsilon_{offset}}{\varepsilon_w}\right]} + 1\right)}\varepsilon_s + \left(\frac{\log[\emptyset]}{\log\left[\left((1-\emptyset)\left(\frac{\varepsilon_s}{\varepsilon_w}\right)^{-0.46cc+0.71} + \emptyset\right)^{-0.46cc+0.71} - \frac{\varepsilon_{offset}}{\varepsilon_w}\right]}\right)}\varepsilon(p)$$

fitting PPMs, as in Wunderlich et al. (2013). Alternatively, it is also frequently fixed as 0.3 assuming spherical solid particles (as in pure sand) (Guillemoteau et al., 2012) or calculated by (Brovelli and Cassiani, 2011):

$$\varepsilon(p=0) = \varepsilon_p; \varepsilon(p=1) = \varepsilon_b \tag{27}$$



**Fig. 3.** Locations of the study sites. Background shows dominant surface texture (European Soil Database v2.0, 2004). Colors represent the texture of the sites: sandy in yellowish, silty in blueish and clayey in reddish. USDA texture triangle showing the particle size distribution categorized by sampling site. The samples presented in Table 2 are represented by triangles. Colors represent the texture of the sites: sandy in yellowish, silty in blueish and clayey in reddish.



Fig. 4. HydraProbe field and lab experiments. A: measuring on a soil vertical profile. B: measuring on a soil sample placed in a PVC tube while it dries down.

These new PPMs have the advantage of including soil texture, for the case that clay is inversely proportional to  $\varepsilon_b$  (i.e., frequencies above  $f_{co}$ ). Lastly, because PPMs of Eq. (25), 26, and 27 do not depend on geometrical parameters, these can be populated without fitting them to calibration data.

# 3. Methodology

# 3.1. Study area, sampled soils and field measurements

In this study, 59 samples were taken across a wide range of soil types at 10 study sites in Belgium, the Netherlands and Serbia (Fig. 3). Soil texture ranged from sand to clay (Fig. 4). At each site, a test pit was dug to identify and sample different soil horizons (i.e., soil layer with distinct physical, biological, chemical and genetic properties) down to the C-horizon (1.3 m to 1.5 m deep). Per soil horizon, a 100-cm<sup>3</sup> intact

undisturbed ring sample was taken, and oven-dried for 24 h at 105 °C to determine  $\theta$  and  $b_d$  (Smith, 2000). Measurements of  $\varepsilon_b$  and T were taken by inserting a HydraProbe sensor (Stevens, Water Monitoring Systems) horizontally into a profile wall (Fig. 5) located 5 to 20 cm away from the sample's extraction point, with the expectation of encountering similar conditions.

# 3.2. Dielectric permittivity measurements

Laboratory and field observations of  $e_b$  and T were all collected using a HydraProbe sensor. HydraProbe characterizes the ratio of the amplitudes of reflected radio waves at 50 MHz with a coaxial waveguide (Logsdon et al., 2010). First, the complex impedance of the soil is calculated by a numerical solution of Maxwell's equations, after which  $e_b^*$ (Eq. (1) is determined (Seyfried et al., 2005; Campbell, 1990).



Fig. 5. Comparison of  $\varepsilon_s$  on soil samples.  $\varepsilon_s$  was calculated using Eq. (14), where  $\varepsilon_b$  was measured with HydraProbe after oven-drying the samples at 105 °C and 150 °C for 24 h. Darker dots show higher soil bound water contents.

 Table 1

 Most useful abbreviations and symbols.

$\sigma_b$	Soil bulk real electrical conductivity
$\varepsilon_b$	Soil bulk real relative dielectric permittivity
θ	Soil volumetric water content
CEC	cation exchange capacity
$b_d$	Soil dry bulk density
Ø	Soil porosity
EM	Electromagnetic
PPM	Pedophysical permittivity model
PTF	Pedotransfer function
$\varepsilon_{h}^{*}$	Soil bulk complex relative dielectric permittivity
$\varepsilon_{b}^{"}$	Soil bulk imaginary relative dielectric permittivity
Eap	Soil bulk apparent relative dielectric permittivity
org	Soil organic matter content
cc	Soil clay content
LR	Lichtenecker and Rother
α	Alpha geometrical parameter
$\varepsilon_w$	Soil water phase real relative dielectric permittivity
$\varepsilon_s$	Soil solid phase real relative dielectric permittivity
$\varepsilon_a$	Soil air phase real relative dielectric permittivity
CRIM	Complex refractive index model
S	Soil saturation
m	Cementation factor
n	Saturation factor
$L_w$	Soil water phase depolarization factor
L	Soil solid phase depolarization factor
$\varepsilon_p$	Bulk real relative dielectric permittivity of an air-water mix
M-W	Maxwell-Wagner
$f_{co}$	Crossover frequency
Т	Soil temperature
SSA	Soil specific surface area
$\theta_{bw}$	Soil volumetric bound water content
$\varepsilon_{bw}$	Soil bound water phase real relative dielectric permittivity
$p_d$	Soil particle (solid phase) density
$\varepsilon_{offset}$	$arepsilon_b$ when $\sigma_b~=0$

Impedance-based sensors are designed to provide the soil water content. In HydraProbe this is derived using an empirical PPM provided by the manufacturer:

$$\theta = 0.109\sqrt{\varepsilon_b} - 0.179\tag{28}$$

Because differences in texture, salinity, bulk density, and *CEC* are not included, the oversimplified PPM of Eq. (28) only provides an indicative water content.

Finally, despite Bosch (2004) warnings about potential variability between individual sensor probes, comprehensive studies (e.g., Seyfried and Murdock 2004; Ojo et al. 2015) have illustrated that HydraProbe's intersensory variability in soil bulk permittivity is negligible and below 1% for water permittivity. Thus, one probe was used for all  $\varepsilon_b$  and *T* data collection.

# 3.3. Water calibration-curve experiment

To test PPMs in soils ranging from dry to nearly saturated conditions,  $\varepsilon_b$  data was obtained from calibration-curve experiments in laboratory. These experiments were conducted using a HydraProbe on ten (repacked) soil samples (see Table 2). These ten soil samples were selected among the 59 samples presented in Fig. 3 (symbolized by triangles), following the criteria to cover a wide range of soil properties.

Laboratory calibration curve experiments imply measuring  $\varepsilon_b$  in a soil sample which water content is changed in controlled conditions. Either, it starts with the soil at saturation point and dries down (Bosch, 2004; Ojo et al., 2015), or the dry soil is increasingly saturated (Le, 2018; Mendieta et al., 2021; Wunderlich et al., 2013). Our saturation experiment in 10 to 14 discrete steps resulted in noisy data because the HydraProbe was reintroduced after each step, then this data was discarded. Therefore, the dry-down approach was adopted.

Before the experiment, the soil samples were oven dried at 105 °C for 24 h to remove all free water (Smith, 2000). A known volume of distilled water was then added to the soil which was thoroughly mixed by hand in a plastic container. The amount of water used was equal to the pore volume for a given bulk volume of soil as measured from the undisturbed soil samples taken in the field, which gave the initial volumetric water content ( $\theta_{init}$ ). A PVC tube of 100 cm<sup>3</sup> closed at its bottom with a perforated plastic cap and filter paper (allowing water to leak) was used as a sampler. The tube was then packed gradually by adding small amounts of mixed soil in increments of 1 cm according to the soil's bulk density. A HydraProbe was then vertically inserted from the top in the filled PVC tube, which was finally placed on an automatic balance with a precision of 0.001 g (see Fig. 4 B). The sample was then allowed to evaporate and leak water. Two times a day, the balance recorded the decrease in total weight ( $w_i$ ) of the sample,  $\varepsilon_h^*$  and *T* were measured with the HydraProbe. At every step (i),  $\theta$  was calculated by Eq. (29).

$$\theta_{i} = \theta_{i-1} - \frac{\frac{(w_{i} - w_{i-1})}{w_{i}} density}{Sampler volume}; \theta_{0} = \theta_{init}$$
<sup>(29)</sup>

Samples did not dry out any further at a certain final step *f*, after which the final water content ( $\theta_f$ ) (i.e. when its weight remained constant; after about 1 to 4 weeks depending on the soil type) was determined, and the experiment was stopped. Samples were then oven dried at 105 °C for 24 h to obtain the remaining free water content ( $\theta_{rem}$ ). The difference between  $\theta_{rem}$  and  $\theta_f$  was used to correct the calibration curves. The correction was applied to the last ten steps of the calibration curves using decreasing weights. Thus, after correction, the final step of the curve is equal to  $\theta_{rem}$ , and  $\theta_{j-k}^{corr}=\theta_{j-k} + (\theta_{rem}-\theta_f)^*(1-0.1^*k)$ , with *k* from 0 to 10. Sample temperature varied between 20 to 24 °C during the measurements, and no correction was applied for this.

# 3.4. Solid phase permittivity and bound water content determination

The solid phase permittivity and bound water content were determined on all 59 samples using the two-phase method (section 2.3.2). First, the samples were oven-dried at 105 °C for 24 h and weighted, and  $\varepsilon_b$  was measured using the HydraProbe. The samples were then oven dried at 150 °C for 24 h to remove the bound water (Smith, 2000) and weighted, after which  $\varepsilon_b$  was determined again with the HydraProbe. The difference in weight of samples dried at 105 and 150 °C allowed to determine the bound water content ( $\theta_{bw}$ ). The permittivity of the solids  $\varepsilon_s$  of samples dried at 105 and 150 °C was determined from Eq. (14)

#### Table 2

Properties of soil samples selected for calibration curve experiments. These were chosen between eight explored sites and following the criteria to cover a wide range of properties. Soil properties were obtained following Smith (2000).

Sample name	Texture class (USDA)	Bulk density [g/cm <sup>3</sup> ]	Clay content (< 2 μm) [g/100 g]	Silt content (50 to 2 µm) [g/100 g]	Sand content (2000 to 50 μm) [g/100 g]	<i>CEC</i> [meq/ 100 g]	Organic matter [g/ 100 g]
A_44	Silty ClayLoam	1.45	11.034	79.997	8.969	8.76	0.444
DREN_8	Clay Loam	1.70	33.046	33.431	33.523	21.70	0.796
D34_8	Sand	1.82	2.521	0.3600	97.119	1.6	0.418
EH2_3	Silty Clay	1.41	38.512	46.706	14.782	32.48	1.670
EH2_6	Sandy Loam	1.59	16.967	29.198	53.835	16.01	0.314
E_44	Loam	1.60	23.233	41.487	35.280	9.61	1.012
HULD_586	Silty Loam	1.60	14.7	78.1	7.2	7.84	0.436
P_17	Loam	1.58	10.980	43.162	45.858	2.42	0.968
VALTHE_N5	Sand	1.58	3.238	2.159	94.604	1.6	0.244
VALTHE_A11	Sand	1.56	3.592	2.515	93.893	1.6	0.334



Fig. 6. Flowchart showing the methodology of PPM development. Colored boxes represent new contributions made by this study.



**Fig. 7.** Results of  $\theta_{bw}$  and  $\varepsilon_s$  experiments using all collected samples (59). Solid phase permittivity  $\varepsilon_s$ , clay content and  $\theta_{bw}$  are correlated showing tree different groups (differentiated by colors) of sandy, silty and clayey samples (yellowish, bluish and reddish, respectively).

following Kameyama and Miyamoto (2008). In Fig. 5,  $\varepsilon_s$  calculated from  $\varepsilon_b$  taken on both types of samples is shown. This illustrates the importance of drying at 150 °C and reveals the largest difference on  $\varepsilon_s$  for samples with high  $\theta_{bw}$ .

# 3.5. Model tests, developments and validations

Similarly to the development of PPMs for the frequencies above  $f_{co}$  (section 2.5), new PPMs are developed for 50 MHz (see flow chart presented in Fig. 6). First, existing volumetric mixing and embedding scheme PPMs (Eq. (5), 7, 9, 11, A.8, and A.10) were, through their geometrical parameters, fitted to laboratory  $\varepsilon_b$  data (Section 3.3). This fitting was performed using different values of geometrical parameters, whereby the optimal value for  $m, \alpha, L$  and  $L_w$  was selected by minimizing the fitting error for each evaluated sample. Second, the obtained geometrical parameter data were linked empirically with clay and *CEC*, creating new empirical PTFs. Finally, these PTFs were combined with existing PPMs to create pre-fitted PPMs (Eq. (36) to (41).

After model development, newly generated PTFs and PPMs were evaluated. For frequencies above  $f_{co}$ , new and existing PPMs were compared (section 4.2). Using collected field and laboratory data at 50 MHz, new pre-fitted PPMs were tested (section 4.3 and 4.4). The developed theoretical PTFs were tested using the obtained geometrical parameters data (section 4.5). Finally, a sensitivity analysis was performed on the best performing pre-fitted PPM (section 4.6).

# 3.6. Sensitivity analysis

Evaluating the influence of soil variables on PPMs is crucial to provide insight into the relationship between soil properties and  $\varepsilon_b$ . For this purpose, after finding the most accurate PPM at 50 MHz, a sensitivity analyses was performed through Monte Carlo simulation. To this end, PPMs were evaluated by iteratively incorporating 100,000 synthetic samples. To ensure evaluating the PPM in a wide range of soil conditions, synthetic data were generated by randomly assigning a value to each of the PPM's variables. While the range of the possible values of each variable is similar to the observed in the collected field data.

#### 3.7. Open code to reproduce all results

To allow reproducing the developed methods and produced results, all soil data and results presented in this work are made available to the reader. Python code compiling all used PPMs and PTFs is available on GitHub (Mendoza Veirana, 2023), and is equally integrated into a



**Fig. 9.** Data obtained from water calibration-curve (dry-dawn) experiments using the ten samples described in Table 2 and Fig. 3 (triangles).  $\varepsilon_{\rm b}$  was measured at 50 MHz using a HydraProbe sensor. The color pallet suggests that they higher the clay content of the sample, the higher  $\varepsilon_{\rm b}$ .

Jupyter Notebook that allows reproducing results and figures incorporated in this paper.

#### 4. Results

# 4.1. Bound water content and solid phase permittivity experiment results

The bound water content  $\Theta_{bw}$  ranged from 0.002 to 0.018%, a positive correlation (0.85) with clay content can be observed in three groups corresponding to a sandy, silty and clayey texture (Fig. 7A). The  $\varepsilon_s$  of all samples generally increased with clay content from 3.3 to 4.1 (Fig. 7B), with an average value of 3.67 (±0.26) (correlation equal to 0.69). Lastly, when plotting  $\theta_{bw}$  in function of  $\varepsilon_s$ , a more disperse plot shows lower correlation (0.52) between both properties (Fig. 7C).



**Fig. 8.** Synthetic evaluation of empirical PPMs (Hallikainen et al., 1985; Jacobsen and Schjønning, 1993 (Eq. (4)) and new modified PPMs (LR (Eq. (25), Linde (Eq. (26), and Endres-Redman (Eq. (27)) at frequencies above  $f_{co}$ . A:  $\varepsilon_b$  in function of clay content (with  $\theta$ =20%). B: predicted  $\varepsilon_b$  using new PPMs in function of  $\theta$ , with clay content = 0 and 40 % (triangles and plus markers, respectively). Additional parameters were fixed as:  $\varepsilon_a = 1$ ,  $\varepsilon_s = 4$ ,  $\varepsilon_w = 80$ ,  $\varepsilon_{offset} = 2.5$ ,  $b_d = 1.4$  g/cm<sup>3</sup> and  $p_d = 2.65$  g/cm<sup>3</sup>.



**Fig. 10.** Using  $\varepsilon_b$  data from calibration curves obtained at 50 MHz (Fig. 9), optimal  $\alpha$  and m were determined by fitting of the LR (panel A and B) and Linde (panel C and D) PPMs (Eq. (5) and (7), respectively). Plots illustrate the relationship between  $\alpha$  and m with respect to the CEC and clay content of the calibrated soil samples, along with the new empirical PTFs (Eq. (30) and (31) for CEC).



**Fig. 11.** Using  $\varepsilon_b$  data from calibration curves obtained at 50 MHz (Fig. 9), optimal depolarization factors *L* and L<sub>w</sub> were determined by fitting of Wunderlich (panel A), Endres-Redman (panel B), Sen (panel C), and Feng-Sen (panel D) PPMs (Eq. (9), 11, A.8 and A.10, respectively). Plots illustrate the relationship between *L* and L<sub>w</sub> with respect to the CEC and clay content of the calibrated soils samples, along with the new empirical PTFs (Eq. (32), 33, 34 and 35).

# 4.2. PPMs evaluation at frequencies abovef co

For frequencies above  $f_{co}$ , new PPMs (Eq. (25), 26 and 27) are compared with the empirical PPMs proposed by Hallikainen et al. (1985), and Jacobsen and Schjønning (1993) (Eq. (4) for varying soil clay content (Fig. 8A), and volumetric water content (Fig. 8B). Overall, for frequencies above  $f_{co}$ , the link between clay content and  $\varepsilon_b$  resulted inversely proportional (Fig. 8A). This is, sandy soils (0% clay content) show higher  $\varepsilon_b$  at given soil–water contents (Fig. 8B).

## 4.3. PPMs test and development at 50 MHz

To illustrate the variability of  $\varepsilon_b$  at 50 MHz, Fig. 9 presents the data of calibration curves for the ten soils described in Table 2, which followed the protocol described in Section 3.2. The curves were corrected with 0 to 4 g of water content. As it was expected at frequencies below  $f_{co}$ , clayey soils show higher  $\varepsilon_b$  than sandy soils at given soil–water contents.

The fitting of volumetric mixing and embedding scheme PPMs (Eq. (5), 7, 9, 11, A.8, and A.10) using the data of calibration curves (Fig. 9) resulted in optimal geometrical parameters. In this way,  $\alpha$  and m were

obtained (See Fig. 10), showing good correlation with the *CEC*, even more than the clay content, as well as *L* and  $L_w$  (see Fig. 11). Thus, we introduce new empirical PTFs at 50 MHz that link the *CEC* with the geometric parameters (Eq. (30) to (35).

$$\alpha = 0.271 \log \left[ CEC \frac{100g}{meq} \right] + 0.306 \tag{30}$$

$$m = n = -0.269 \log \left[ CEC \frac{100g}{meq} \right] + 1.716$$
(31)

$$L_w = -0.0493 \log \left[ CEC \frac{100g}{meq} \right] + 0.1279$$
(32)

$$L = -0.194 \log \left[ CEC \frac{100g}{meq} \right] + 0.472$$
(33)

$$L = -0.186\log\left[CEC\frac{100g}{meq}\right] + 0.565$$
(34)

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$$L = -0.193 \log \left[ CEC \frac{100g}{meq} \right] + 0.44 \tag{35}$$

To propose new optimized PPMs at 50 MHz that do not require calibration data, the new PTFs (Eq. (30) to (35) are replaced in volumetric mixing and embedding schemes, resulting in new pre-fitted PPMs:

If in LR PPM (Eq. (5),  $\alpha$  is calculated by Eq. (30), this becomes:

$$\varepsilon_{b} = \theta^{-0.269\log\left[CEC\frac{100g}{meq}\right] + 1.716} (\varepsilon_{w} - \varepsilon_{a}) + \emptyset^{-0.269\log\left[CEC\frac{100g}{meq}\right] + 1.716} (\varepsilon_{a} - \varepsilon_{s}) + \varepsilon_{s}$$
(37)

If in Wunderlich PPM (Eq. (9),  $L_w$  is calculated by Eq. (32), this becomes:

$$\varepsilon_{b} = \begin{pmatrix} 0.271 \log \left[ CEC \frac{100g}{meq} \right] + 0.306 & 0.271 \log \left[ CEC \frac{100g}{meq} \right] + 0.306 & 0.271 \log \left[ CEC \frac{100g}{meq} \right] + 0.306 \\ + (\mathcal{Q} - \theta)\varepsilon_{a} & + (\mathcal{Q} - \theta)\varepsilon_{a} \end{pmatrix}^{1/\left(0.271 \log \left[ CEC \frac{100g}{meq} \right] + 0.306 \right)}$$
(36)

If in Linde PPM (Eq. (7), m and n is calculated by Eq. (31), this becomes:

$$\frac{d\varepsilon(p)}{dp} = \frac{\varepsilon(p)(\theta_n - \theta_1)}{1 + \theta_1 - \theta_n + p(\theta_n - \theta_1)}$$

$$\frac{\varepsilon_w - \varepsilon(p)}{\left(-0.0493\log[CEC\frac{100g}{m}] + 0.1279\right)\varepsilon_w + \left(0.0493\log[CEC\frac{100g}{m}] + 0.8721\right)\varepsilon(p)}$$



Fig. 12. PPM's RMSE (top) and R2 (bottom) after evaluation on the data of calibration curves (Fig. 9) obtained at 50 MHz (data in Table A2).



Fig. 13. Observed  $\varepsilon_{\rm b}$  data were obtained in 10 vertical soil profiles next (5 to 20 cm) to the extracted samples (59) using a HydraProbe sensor (section 3.1). These observations are compared to predicted  $\epsilon_{b}$  using LR, Linde and Endres-Redman PPMs (from top to bottom, respectively). Left panels (A, C, E) present the fixed-parameter PPMs, while the right panels (B, D, F) present the newly developed PPMs. Bigger dots represent independent observations (i.e., from sampled sites not used to develop the new PPMs).

$$\varepsilon(p=0) = \varepsilon_1; \varepsilon(p=1) = \varepsilon_b$$
 (38)

If in Endres-Redman PPM (Eq. (11), L is calculated by Eq. (33), this becomes:

$$\begin{split} \frac{d\varepsilon(p)}{dp} &= \frac{\varepsilon(p)(1-S)}{S+p(1-S)} \\ \frac{\varepsilon_a - \varepsilon(p)}{\left(-0.194 \log\left[CEC\frac{100g}{meq}\right] + 0.472\right)\varepsilon_a + \left(0.194 \log\left[CEC\frac{100g}{meq}\right] + 0.528\right)\varepsilon(p)} \\ \varepsilon(p=0) &= \varepsilon_w; \varepsilon(p=1) = \varepsilon_p \end{split}$$

$$\frac{d\varepsilon(p)}{dp} = \frac{\varepsilon(p)(1-\emptyset)}{\emptyset+p(1-\emptyset)}$$

$$\frac{\varepsilon_s - \varepsilon(p)}{\left(-0.194\log\left[CEC\frac{100g}{meq}\right] + 0.472\right)\varepsilon_s + \left(0.194\log\left[CEC\frac{100g}{meq}\right] + 0.528\right)\varepsilon(p)}$$

$$\varepsilon(p=0) = \varepsilon_p; \varepsilon(p=1) = \varepsilon_b$$
(39)

If in Sen PPM (Eq. A.8), L is calculated by Eq. (34), this becomes:

$$\begin{aligned} \frac{d\varepsilon(p)}{dp} = & \frac{\varepsilon(p)(1 - \emptyset + \theta)}{(\emptyset + \theta) + p(1 - \emptyset + \theta)} \\ \frac{\varepsilon_{cg} - \varepsilon(p)}{\left(-0.186\log\left[CEC\frac{100g}{meq}\right] + 0.565\right)\varepsilon_{cg} + \left(0.186\log\left[CEC\frac{100g}{meq}\right] + 0.435\right)\varepsilon(p)} \end{aligned}$$

 $\varepsilon_{cg} = \varepsilon_{w} \frac{(1 - \emptyset)\varepsilon_{s} + \theta \left[ \left( -0.186 \log \left[ CEC \frac{100g}{meq} \right] + 0.565 \right) \varepsilon_{s} + \left( 0.186 \log \left[ CEC \frac{100g}{meq} \right] + 0.435 \right) \varepsilon_{w} \right]}{(1 - \emptyset)\varepsilon_{w} + \theta \left[ \left( -0.186 \log \left[ CEC \frac{100g}{meq} \right] + 0.565 \right) \varepsilon_{s} + \left( 0.186 \log \left[ CEC \frac{100g}{meq} \right] + 0.435 \right) \varepsilon_{w} \right]}$ 

1 /



Fig. 14. Diamond marks show  $\alpha$ , m and L data presented in Section 4.3, these are obtained by fitting of PPMs (Eq. (5), 7 and 11, respectively) on data of calibration curves (Fig. 9) at 50 MHz. Theoretical PTFs were evaluated to predict m (upper) and L (lower) in function of  $\alpha$ . Eq. (20) was evaluated with  $\varepsilon_s = 3.7$ ,  $\varepsilon_w = 80$  and  $\varepsilon_{offset} = 2.5$ .

$$\varepsilon(p=0) = \varepsilon_a; \varepsilon(p=1) = \varepsilon_b \tag{40}$$

If in Feng-Sen PPM (Eq. A.10), L is calculated by Eq. (35), this becomes:

$$\begin{aligned} \frac{d\varepsilon(p)}{dp} &= \frac{\varepsilon(p)}{\theta + p(1-\theta)} \left[ (1-\emptyset) \\ \frac{\varepsilon_s - \varepsilon(p)}{\left( -0.193 \log \left[ CEC \frac{100g}{meq} \right] + 0.44 \right) \varepsilon_s + \left( 0.193 \log \left[ CEC \frac{100g}{meq} \right] + 0.56 \right) \varepsilon(p)} + (\emptyset - \theta) \\ \frac{\varepsilon_a - \varepsilon(p)}{\left( -0.193 \log \left[ CEC \frac{100g}{meq} \right] + 0.44 \right) \varepsilon_a + \left( 0.193 \log \left[ CEC \frac{100g}{meq} \right] + 0.56 \right) \varepsilon(p)} \right] \end{aligned}$$

$$\varepsilon(p=0) = \varepsilon_w; \varepsilon(p=1) = \varepsilon_b \tag{41}$$

The new PPMs of Eq. (36) to (41) differs on those of Eq. (25) to (27) because they are developed for EM frequencies below and above  $f_{co}$ , respectively. Subsequently, all PPMs (including the list in Table A1) were tested using the data of calibration curves presented in section 3.2 (Fig. 9). Two error indicators were used: the root mean square error (RMSE) and coefficient of determination ( $R^2$ ) (Table A2, Fig. 12). Based on the nature of the PPM and its evaluation, these were split in four groups: empirical models, volumetric mixing and embedding schemes

models with fixed geometric parameters, volumetric mixing and embedding schemes models fitted to  $\varepsilon_b$  using geometrical parameters, and new pre-fitted PPMs.

#### 4.4. Fieldwork results

The newly developed PPMs were compared with their fixedparameter counterparts using field data obtained in soil profiles (section 3.1) (see Fig. 13). Samples collected in two of these soil profiles ('S', 'Hoeke'), were not used in the development of the new PPMs, as these were reserved for independent validation.

# 4.5. Evaluation of theoretical pedotransfer functions

New and well-established PTFs that link geometrical parameters, are tested using data obtained at 50 MHz (Fig. 14). The new theoretical PTF (Eq. (20) improved prediction of m ( $R^2 = 0.99$ ) compared to the classical formula  $m = 1/\alpha$  (Eq. (18) ( $R^2 = -1.17$ ), particularly when  $\alpha < 0.8$ . To calculate *L* in function of  $\alpha$ , a combination of Eq. (23) with 20 is more convenient.

# 4.6. Sensitivity analysis

The significance of various soil properties was evaluated using a Monte Carlo simulation applied to the new LR PPM (Eq.36) developed for 50 MHz. Hereby, 100,000 randomly generated samples were introduced into Eq. (36). Fig. 15 shows the result of this simulation, where each blue dot represents the predicted  $\varepsilon_b$  of a synthetic sample.



Fig. 15. Sensitivity analysis result of the new LR PPM (Eq. (36) for 50 MHz. The evaluation consisted on a Monte Carlo simulation where the PPM was repeatedly evaluated with 100,000 randomly generated virtual samples.  $\varepsilon_a = 1.3$  was fixed.

The observed range of soil properties indicates that the predicted  $\varepsilon_b$  remains largely unaffected by variations in  $b_d$ ,  $p_d$  and  $\varepsilon_s$ . The soil property  $\varepsilon_w$  shows slight influence on  $\varepsilon_b$ . Soil *CEC* emerges as the second most impactful property. For instance, samples with *CEC* at 1.6 meq/100 g correspond to a  $\varepsilon_b$  range from 5 to 40, whereas higher *CEC* values (= 40 meq/100 g) yield an  $\varepsilon_b$  range from 10 to 55. Finally, as anticipated, water content influences  $\varepsilon_b$  the most. Dry field samples with  $\theta = 5\%$  correspond to a  $\varepsilon_b$  range from 5 to 10. Conversely, wet samples with  $\theta = 50\%$  reach a  $\varepsilon_b$  range from 30 to 50, regardless of any other soil property.

#### 5. Discussion and limitations

For frequencies above  $f_{co}$ , Fig. 2 show good agreement between new and well-established PTFs. However, it is worth mentioning that empirical PTFs are based on data correlations, and do not necessarily have a physical basis. Despite these PTFs were developed using different datasets presented by different authors (Section 2.4), their integration into well-stablished PPMs resulted on in-accordance  $\varepsilon_b$  predictions (Fig. 8). This approach allowed to make the prediction of  $\varepsilon_b$  sensitive to clay content. New PPMs (Eq. (25), 26 and 27) evaluation show a reduction in  $\varepsilon_b$  values when clay content increases (Fig. 8). This suggest that at frequencies above  $f_{co}$ , where the M-W affect is minor, the water bounded to clay particles reduces  $\varepsilon_b$  (Cassidy and Jol, 2009).

Despite PPMs were tested using new laboratory  $\varepsilon_b$  data, some

empirical PPMs are expressed in function of  $\varepsilon_{ap}$  (Eq. (3), 4, A.1, A.2, A.3, and A.4). While it is common not to differentiating between  $\varepsilon_b$  and  $\varepsilon_{ap}$ since they are often considered interchangeable in PPM testing (Wunderlich et al., 2013; Steelman and Endres, 2011), it is important to note that integrating  $\varepsilon_{a\!p}$  resulted in poor performance. Therefore, this can be seen as a limitation. After the PPM test, the four classes showed different accuracies (Fig. 12). Surprisingly, empirical, fixed-parameter volumetric mixing and embedding scheme PPMs presented similarly high errors, which reach negative  $R^2$  on clayey and sandy samples (see Table A2). This is probably due to the poor flexibility of such models in capturing influences of soil characteristics such as CEC or soil texture on lowfrequency  $\varepsilon_b$  observations. As expected, the fitted PPMs, particularly Eq. (9) (Wunderlich et al., 2013), were the most accurate. Ultimately, the new pre-fitted PPMs provide a significant improvement compared to the fixed parameter PPMs. For example, the average prediction error of the new Linde PPM (Eq. (37) over all tested samples (RMSE = 1.56 was reduced by 412% compared to the fixed parameter version (Eq. (8), RMSE = 6.43).

The new empirical PTFs for 50 MHz (Eq. (30) to (35) seem to show inconsistencies due to  $\alpha > 1$  (see e. g. Ponizovsky et al., 1999) and L < 0 values (Figs. 10 and 11), which has no physical basis in the context of volumetric mixing and embedding scheme theory. This is probably because Eq. (30) to (35) are based on empirical correlations. Particularly, in the PTF of Eq. (34) *L* is negative for *CEC* > 25 meq/100 g. Then, its integration in the new PPM of Eq. (40) was restricted to positive

values, otherwise the Eq. (38) diverged.

Estimations of  $\varepsilon_s$  at 50 MHz show an average of 3.67, which is higher in clayey than in sandy soils, possibly due to small remains of  $\theta_{bw}$  in the samples, even after drying at 150 °C for 24 h. Possible weight reduction due to organic matter burned was dismissed. Despite acquiring  $\theta_{bw}$ , its introduction in Eq. A.7 (Dobson et al., 1985) was not effective to obtain  $\varepsilon_{bw}$  by fitting process. In fact, totally unrealistic  $\varepsilon_{bw}$  values (-5000 to 1700) were reached, and correlation with *CEC* or clay content was insignificant. The Dobson PPM (Eq. A.7) results in higher errors than other fitting PPMs, probably because it was originally developed for frequencies above  $f_{co}$ ; then no new models are proposed.

Based on the soil *CEC*, shrinking-swelling and non-expanding clays cannot be differentiated, which could limit PPM usage, because crack development (observed in the EH2\_6 and DREN\_8 samples) affects  $e_b$  readings. Additionally, *CEC* values < 1.6 meq/100 g were unmeasurable, then some uncertainty remains in Eq. (30) to (35) for this *CEC* range (Figs. 10 and 11).

At frequencies above  $f_{co}$ , increased *T* and water salinity reduces  $\varepsilon_b$  (Fig. 1). This effect is normally accounted for by using models similar to Eq. (13). However, at 50 MHz (frequency below  $f_{co}$ ), this correction was not satisfactory because the effect is opposite; since temperature influences  $\sigma_b$  positively, and thus  $\varepsilon_b$  (via M-W effect) (e.g., Chen and Or, 2006a).

For the field data test at 50 MHz, the prediction accuracy of the new PPMs is significantly higher than that of the fixed-parameter PPMs, even when considering the independent field data from the 'S' and 'Hoeke' sites. The remaining errors of new PPMs, mostly variance errors, can be partly attributed to the distance between the extraction point of the soil cores and the HydraProbe measurements (5–20 cm), as well as differences in temperature, salinity and soil structure. It is worth noting is that well-established fixed-parameter PPMs, such as the CRIM and Linde model, resulted in negative  $R^2$  values. This is probably due to their incapability of consider soil texture or *CEC*, and that  $\varepsilon_b$  is observed at a frequency below  $f_{co}$ , where the M-W effect has to be considered.

Importantly, our field results showed a high  $R^2$  (=0.92) for the laboratory-based Endres-Redman PPM (Eq. (39), despite several authors (i.e., Bosch, 2004; Ojo et al., 2015; Vaz et al., 2013) recommending separate laboratory and in-situ field model calibrations. The difference between the data collected at laboratory and in field conditions, is that the latter was obtained from samples presenting soil structure and a larger temperature range (11 to 30 °*C*). However, the new Endres-Redman PPM of Eq. (39) performed with  $R^2$  (=0.93) for laboratory data, suggesting that soil structure and field temperatures influenced the 1% of  $\varepsilon_b$  prediction.

The test of well-stablished and new theoretical PTFs using data at 50 MHz (section 4.5) show novel results. While the new PTF (Eq. (20) is a general case of  $m = 1/\alpha$ , its significant improvement in accuracy can be attributed to the assumptions introduced in section 2.4, which agree with the volumetric mixing theory (e.g., Roth et al., 1990). Both PTFs assume Archie's law and that  $\varepsilon_b$  is as in LR PPM (Eq. (5) for saturated conditions, even though they are applied in soils with saturation ranging from dry to fully saturated. Expanding on Brovelli and Cassiani (2008), the newly developed PTF (Eq. (20) also assumes  $\varepsilon_s \neq 0$ , that implies  $\varepsilon_{\textit{offset}} 
eq 0$ . This is derived from CRIM model for dry conditions (Eq. (14), which states that with a water content of zero,  $\varepsilon_b$  is nonnegligible when  $\varepsilon_s \neq 0$ . Considering that  $\sigma_b = 0$  with zero water content (Archie's law), this further implies that the  $\varepsilon_{\textit{offset}}$  considered by Hilhorst (Hilhorst, 2000), and presented in Eq. (19), cannot be ignored either. In summary, the proposed PTF (Eq. (20) assume that a soil's geometrical arrangement is independent from  $\theta$  (as well as  $m = 1/\alpha$ ), but also that  $\varepsilon_s$  (and therefore  $\varepsilon_{\textit{offset}}$  ) has to be accounted for.

Finally, the sensitivity analysis illustrate that as expected, the volumetric water content is the most influential variable in  $\varepsilon_b$  prediction.

This is followed by *CEC* (in agreement with Ojo et al., 2015) and  $\varepsilon_w$ . Variations of  $b_d$ ,  $p_d$  and  $\varepsilon_s$  over the observed range are irrelevant in the calculation of  $\varepsilon_b$ , suggesting that at frequencies below  $f_{co}$  these could be fixed as 1.5 g/cm<sup>3</sup>, 2.65 g/cm<sup>3</sup>, and 3.7, respectively.

# 6. Conclusions

For the first time, the widely acknowledged dependence of  $\varepsilon_b$  on soil texture and *CEC* is included in PPMs through the geometric parameters for both frequencies above and below  $f_{co}$ . Furthermore, newly modified PPMs are proposed for frequencies above  $f_{co}$ , showing good agreement with prior observations. For frequencies below  $f_{co}$ , geometrical parameters are optimally correlated with soil *CEC*.

The relationship between geometrical parameters using the classic pedotransfer function  $m = 1/\alpha$  ( $R^2$ =-1.17) was significantly improved by a new theoretical PTF ( $R^2$ =0.99). This improvement is because  $\varepsilon_s$  and  $\varepsilon_{offset}$  are accounted for. Additionally, the new PTF is easy to implement, and the lacking of dielectric dispersion assumption allows its applicability for data at all EM frequencies.

Testing of 28 PPMs at 50 MHz shows that all empirical PPMs, and fixed parameter volumetric mixing and embedding schemes, have to be avoided when predicting  $\varepsilon_b$  due to the potentially negative  $R^2$ . This is especially relevant in pure sand, silty-clay and clay samples. Instead, the pre-fitted new PPMs can be implemented in laboratory and field data analysis using  $p_d = 2.65$ ,  $b_d = 1.5$  and  $\varepsilon_s = 3.7$ . These new PPMs are highly accurate (new Linde PPM reduced its RMSE fourfold), grasp the full complexity of soils with regards to EM frequencies, and no calibration data is further needed. However, if  $\varepsilon_b$  is known, the fitting model of Wunderlich et al. (2013) performs best.

The large influence of *CEC* suggests that this could be quantified *in* situ when  $\varepsilon_b$  and  $\theta$  are known. The low influence of  $b_d$  suggests that its variation (e.g., due to soil compaction processes) is unlikely to be detected with low-frequency instrumentation. Similarly, soil structure and temperature variations between 11 to 30 °*C* do not play a major role in  $\varepsilon_b$  at 50 MHz. While further challenges remain, such as accounting for the influence of salinity or cracks in clayey soils, this work provides a comprehensive step towards optimizing the prediction of  $\varepsilon_b$ . Combined with an open workflow, this provides a robust basis to further improve geophysical approaches to soil characterization

#### **Declaration of Competing Interest**

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

# Data availability

All data and code is shared as attached files and online in a Github open repository

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# Declaration of generative AI and AI-assisted technologies in the writing process

During the preparation of this work the author(s) used GPT-4 OpenAI in order to check readability. After using this tool/service, the author(s) reviewed and edited the content as needed and take(s) full responsibility for the content of the publication.

# Appendix A

# Table A1

Pedophysical permittivity models. A coded implementation of these functions is available in the 'Pedophysical permittivity models.py' file supplemented to this paper.  $\varepsilon_{cg}$ : permittivity of coated grains.

Model reference	Equation	number
<i>Empirical models</i> Nadler et al. (1991)	$\theta = -7.25 \bullet 10^{-2} + 3.67 \bullet 10^{-2} \varepsilon_{ap} - 12.3 \bullet 10^{-4} \varepsilon_{ap}^2 + 15 \bullet 10^{-6} \varepsilon_{ap}^3$	A.1
Jacobsen and Schjønning (1993)	$ heta = -7.01 ullet 10^{-2} + 3.47 ullet 10^{-2} arepsilon_{ap} - 11.6 ullet 10^{-4} arepsilon_{ap}^2 + 18 ullet 10^{-6} arepsilon_{ap}^3$	A.2
Malicki et al. (1996)	$arepsilon_{ap} = 0.819 + 0.168 b_d + 0.159 b_d^2 + (7.17 + 1.18 b_d)  heta$	A.3
Steelman and Endres (2011)	$ heta = -0.157 + 5.65 ullet 10^{-2} arepsilon_{ap} - 2.03 ullet 10^{-3} arepsilon_{ap}^2 + 2.97 ullet 10^{-5} arepsilon_{ap}^3$	A.4
Logsdon et al. (2010)	$ heta = 2.2 \bullet 10^{-2} \varepsilon_b - 4.7 \bullet 10^{-4} \varepsilon_b^2 + 5.14 \bullet 10^{-6} \varepsilon_b^3$	A.5
Volumetric mixing models		A.6
Peplinski et al. (1995)	$\varepsilon_b = \left[1 + \frac{b_d}{p_d} \varepsilon_s^r \partial^{\beta} \varepsilon_w^r - \theta\right]^{\overline{\tau}}$ Where $\beta = 1.2748 - 0.519$ s and $-0.152cc; \tau = 0.65$	
Dobson et al. (1985)	$\varepsilon_{b} = \frac{3\varepsilon_{s} + 2(\theta(\varepsilon_{w} - \varepsilon_{s}) + 2\theta_{bw}(\varepsilon_{bw} - \varepsilon_{s}) + 2(\emptyset - \theta)(\varepsilon_{a} - \varepsilon_{s})}{(\varepsilon_{bw} - \varepsilon_{s}) + 2(\theta - \theta)(\varepsilon_{bw} - \varepsilon_{s})}$	A.7
	$3 + \theta \Big( \frac{\varepsilon_s}{\varepsilon_w} - 1 \Big) + \theta_{bw} \Big( \frac{\varepsilon_s}{\varepsilon_{bw}} - 1 \Big) + ( \varnothing - \theta ) \Big( \frac{\varepsilon_s}{\varepsilon_a} - 1 \Big)$	
Embedding schemes	$d\varepsilon(p) = \varepsilon(p)(1-\varphi+\theta) \qquad \varepsilon_{cg}-\varepsilon(p) \qquad (1-\varphi)\varepsilon_s+\theta[L\varepsilon_s+(1-L)\varepsilon_w] = (n-\varphi)\varepsilon_s+\varepsilon(p-1) = \varepsilon_{cg}-\varepsilon(p)$	A.8
Sen et al. (1981)Sen et al. (1981)	$\frac{1}{dp} = \frac{1}{(\omega + \theta) + p(1 - \omega + \theta)} \frac{1}{L\epsilon_{cg} + (1 - L)\epsilon(p)} \epsilon_{cg} = \epsilon_w (1 - \omega)\epsilon_w + \theta[L\epsilon_s + (1 - L)\epsilon_w]} \epsilon(p = 0) = \epsilon_a \cdot \epsilon(p = 1) = \epsilon_b \text{ when } L = 0$	A.9
	0.3	
Feng and Sen (1985)Feng and Sen (1985)	$\frac{d\varepsilon(p)}{dp} = \frac{\varepsilon(p)}{\theta + p(1-\theta)} \bigg[ (1-\varnothing) \frac{\varepsilon_s - \varepsilon(p)}{L\varepsilon_s + (1-L)\varepsilon(p)} + (\varnothing - \theta) \frac{\varepsilon_a - \varepsilon(p)}{L\varepsilon_a + (1-L)\varepsilon(p)} \bigg] \varepsilon(p=0) \\ = \varepsilon_{w}; \varepsilon(p=1) = \varepsilon_b \text{ When } L = 0.3$	A.10 A.11

Table A2		
Root mean square error (RMSE) (upper) and R <sup>2</sup>	$^2$ (lower) between $\epsilon_b$ observed in laboratory calibration curves and PPM's $\epsilon_l$	prediction.

DMOL D2	A 44	DREN 9	D24.0	EU2 2	EU2 6	E 44		D 17	VALTHE NE	VALTHE A11	Moor
KMSE K~	A_44	DKEN_8	D34_8	EE12_3	ЕП2_6	E_44	UULD_280	r_17	VALIHE_N5	VALITE_ATT	mean
Empirical PPMs	6.85	9.24	2.20	12.68	6.15	4.10	6.58	0.80	1.81	1.59	5.20
Topp (Eq. (3)	0.44	-1.52	0.56	-0.33	0.41	0.50	0.39	0.98	0.88	0.93	0.33
Jac-SchjA (Eq. (4)	4.85	8.19	3.29	11.20	4.52	3.18	4.80	2.48	3.65	3.41	4.96
	0.72	-0.98	0.02	-0.04	0.68	0.70	0.68	0.85	0.50	0.66	0.38
HydraProbe (Eq. (25)	4.82	7.76	3.62	10.96	4.35	2.73	4.65	2.17	3.61	3.43	4.81
	0.72	-0.78	-0.19	0.01	0.70	0.78	0.70	0.89	0.51	0.66	0.40
Nadler (Eq. A1)	5.88	8.42	3.49	9.84	5.04	5.19	5.42	6.88	7.16	7.36	6.47
	0.59	-1.09	-0.10	0.20	0.60	0.20	0.59	-0.15	-0.94	-0.56	-0.07
Jac-SchjB (Eq. A2)	4.12	8.18	4.94	12.3	3.77	3.11	3.92	3.27	5.36	5.11	5.41
	0.80	-0.97	-1.21	-0.25	0.78	0.72	0.79	0.74	-0.09	0.25	0.15
Malicki (Eq. A3)	7.43	8.39	3.29	13.56	5.78	3.83	6.04	1.15	2.38	1.98	5.38
	0.35	-1.08	0.02	-0.52	0.48	0.57	0.49	0.97	0.78	0.89	0.29
Steel-Endr (Eq. A4)	9.74	11.10	0.66	14.95	8.51	5.98	9.12	2.16	1.43	1.95	6.56
	-0.13	-2.63	0.96	-0.85	-0.13	0.06	-0.16	0.89	0.92	0.89	-0.03
Logsdon (Eq. A5)	4.66	8.28	3.87	10.72	4.38	3.83	4.59	4.12	4.61	4.53	5.36
	0.74	-1.02	-0.35	0.05	0.70	0.57	0.70	0.59	0.20	0.41	0.26
Fixed parameter PPMs	8.35	9.64	1.58	14.07	7.28	4.71	7.45	1.07	1.14	1.11	5.64
CRIM (Eq. (6)	0.17	-1.74	0.77	-0.64	0.17	0.35	0.22	0.97	0.95	0.96	0.22
Linde (Eq. (8)	9.72	11.00	0.87	15.47	8.40	6.04	8.65	2.09	1.00	1.10	6.43
	-0.12	-2.57	0.93	-0.98	-0.10	-0.08	-0.05	0.89	0.96	0.96	-0.01
Wunderlich (Eq. (10)	3.09	6.09	5.50	9.46	2.67	2.19	2.85	3.67	5.83	5.66	4.70
	0.89	-0.09	-1.74	0.26	0.89	0.86	0.89	0.67	-0.29	0.08	0.24
Endr-Redm (Eq. (12)	7.09	9.02	2.95	13.06	6.38	4.06	6.53	0.90	1.92	1.65	5.36
	0.40	-1.40	0.21	-0.41	0.36	0.51	0.40	0.98	0.86	0.92	0.28
Peplinski (Eq. A6)	7.95	6.43	10.85	12.61	2.31	1.84	6.94	2.74	11.16	11.10	7.39
	0.25	-0.22	-9.67	-0.32	0.92	0.90	0.32	0.82	-3.72	-2.54	-1.33
Sen (Eq. A9)	6.51	7.33	4.18	12.82	4.69	2.88	4.97	1.76	3.45	3.15	5.17
	0.50	-0.58	-0.58	-0.36	0.66	0.75	0.65	0.92	0.55	0.72	0.32
Feng-Sen (Eq. A11)	8.10	10.25	1.61	14.18	7.44	5.30	7.50	1.97	1.57	0.94	5.89
	0.22	-2.10	0.76	-0.66	0.13	0.17	0.21	0.91	0.91	0.97	0.15
Fitted parameter PPMs	1.17	2.05	0.17	1.05	0.89	1.47	0.89	0.56	0.97	1.10	1.03
LR (Eq. (5)	0.98	0.88	1.00	0.99	0.99	0.94	0.99	0.99	0.96	0.97	0.97
Linde (Eq. (7)	1.18	2.02	0.31	1.01	0.90	1.55	0.90	0.66	0.92	0.98	1.04
	0.98	0.88	0.99	0.99	0.99	0.93	0.99	0.99	0.97	0.97	0.97
Wunderlich (Eq. (9)	1.15	1.17	0.25	0.89	0.95	0.63	0.88	0.41	0.93	0.97	0.82
	0.98	0.96	0.99	0.99	0.99	0.99	0.99	1.00	0.97	0.97	0.98
Endr-Redm (Eq. (11)	1.41	2.23	0.27	3.74	1.13	1.84	1.04	0.90	1.09	1.14	1.48
	0.98	0.85	0.99	0.88	0.98	0.90	0.98	0.98	0.96	0.96	0.95
Dobson (Eq. A7)	1.66	1.41	2.15	2.62	9.51	0.86	1.50	1.23	1.69	1.12	2.38
	0.97	0.94	0.58	0.94	-0.41	0.98	0.97	0.96	0.89	0.96	0.78
Sen (Eg. A8)	1.28	2.53	0.36	4.47	0.83	1.61	0.97	0.94	0.69	0.81	1.45
	0.98	0.81	0.99	0.83	0.99	0.92	0.99	0.98	0.98	0.98	0.95
Feng-Sen (Eq. A10)	2.29	3.31	0.84	5.43	1.89	2.91	2.06	1.72	1.11	0.85	2.24
	0.94	0.68	0.94	0.76	0.94	0.75	0.94	0.93	0.95	0.98	0.88

(continued on next page)

Table A2 (continued)

<b>RMSE</b> $R^2$	A_44	DREN_8	D34_8	EH2_3	EH2_6	E_44	HULD_586	P_17	VALTHE_N5	VALTHE_A11	Mean
New	1.62	2.12	0.87	2.06	2.34	2.28	1.65	0.63	1.03	1.39	1.60
pre-fitted	0.97	0.87	0.93	0.96	0.91	0.85	0.96	0.99	0.96	0.94	0.94
PPMs											
LR (Eq. (36)											
Linde (Eq. (37)	2.31	2.18	0.96	1.06	2.03	1.81	2.44	0.78	0.92	1.08	1.56
	0.94	0.86	0.92	0.99	0.94	0.90	0.92	0.99	0.97	0.97	0.94
Wunderlich (Eq. (38)	3.87	3.78	1.12	6.33	1.56	2.24	3.96	0.85	0.97	1.21	2.59
	0.82	0.58	0.89	0.67	0.96	0.85	0.78	0.98	0.96	0.96	0.85
Endr-Redm (Eq. (39)	1.85	2.30	0.89	3.68	1.77	2.33	1.87	0.90	1.09	1.25	1.79
	0.96	0.84	0.93	0.89	0.95	0.84	0.95	0.98	0.96	0.96	0.93
Sen (Eq. (40)	3.13	2.49	1.62	4.47	1.07	1.73	2.58	0.97	0.69	0.97	1.97
	0.88	0.82	0.76	0.83	0.98	0.91	0.91	0.98	0.98	0.97	0.90
Feng-Sen (Eq. (41)	2.45	3.54	1.05	4.98	2.04	3.06	2.71	1.76	2.11	1.14	2.48
	0.93	0.63	0.90	0.80	0.94	0.72	0.90	0.92	0.83	0.96	0.85

# Appendix B. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.geoderma.2023.116624.

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