Estimation of vineyard soil structure and preferential flow using dye tracer, X-ray tomography, and numerical simulations

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ABSTRACT
The appearance and distribution of soil pores have a significant influence on water flow and solute transport in the soil vadose zone. The pore system is highly variable in arable soils where crop rotation, tillage, trafficking, soil amendments, and various management practices are commonly implemented. The aim of this study was to assess the porous system and preferential flow pathways in a vineyard soil using undisturbed soil columns, and by combining laboratory and numerical methods with dye staining and X-ray imaging. It was hypothesized that the integration of various methods could reveal more information about soil structure, and flow and transport behavior of structured arable soil. Soil water retention and hydraulic conductivity curves were obtained during the evaporation experiments. The transport of Brilliant Blue were simulated using HYDRUS-1D. A single-porosity model of soil hydraulic properties provided a good description of data collected during the evaporation experiments. Data collected during leaching experiments did not provide enough experimental evidence for the occurrence of nonequilibrium flow patterns and the differentiation between the single- and dual-permeability models of soil hydraulic properties. However, dye staining and X-ray imaging revealed a complex pore-architecture network with large vertical and horizontal biopores. The staining patterns (Brilliant Blue FCF) within the vertical column sections documented the extent of preferential flow. The study showed that the bi-modal character of pore structure could often be hidden when a limited number or non-adequate methods are applied for its quantification from water flow behavior. The impact of preferential pathways on dye transport can be investigated with observations and simulations. A combination of various methods enabled us to adequately assess vineyard soil structure and fine-tune the description and extent of preferential water flow.

1. Introduction

Most pronounced soil heterogeneity in terms of structural changes is present in the topsoil layer of arable soils, which are heavily modified during each crop growing season by tillage and trafficking (Jirků et al., 2013; Strudley et al., 2008). Changes in climate, biological activity, and soil management practices may have a strong influence on soil structure (Fér et al., 2016, 2019; Kodešová et al., 2011). Pagliai et al. (2004) demonstrated deterioration of the soil structure in arable soils due to enhanced oxidation of organic matter by aeration, mechanical dispersion because of moist soil compression, or disaggregation of bare soil by rainfall. A newly developed pore system (e.g., after the tillage) in arable soils, even with a well-connected network of larger pores, is often unstable and changes rapidly. As reported by Horn (2004), the temporal variability of soil structure and its evolution is influenced by various biotic and abiotic factors, and it is often challenging to properly estimate its influence on subsurface flow and transport properties. In the Mediterranean, vineyards are one of such agricultural systems with enhanced soil structure degradation due to topography, climatic conditions, intensive management, and trafficking practices (Bogunović et al., 2020; Galati et al., 2015; Prosdocimi et al., 2016a, 2016b). Viticulture areas register some of the highest rates of soil and water losses in Europe as a result of employing unsustainable practices and management (e.g., large interrow widths, low leaf cover, steep slopes, and...
Preferential flow, i.e., unequal or nonequilibrium flow, is a phenomenon in which liquid, gas, and dissolved chemicals primarily move through a small fraction of the porous media, such as wormholes, root holes, and cracks, thus allowing much faster transport (Gerke, 2006; Ranjit Kumar et al., 2017). The most common types of preferential flow are macropore flow (through biopores or erosion cracks), finger flow (unequal water flow due to the hydraulic instability or heterogeneity), and funnel flow (flow above sloping soil layers). At some sites, the combination of different flow patterns can occur as well (Guo and Lin, 2018). Preferential flow is mostly dependent on pore characteristics and soil properties, especially soil texture and structure, as well as on hydraulic properties, and the human impact through soil management practices in agricultural soils. Macropores have a positive influence on preferential flow due to their number, size, linkage, and continuity (Jarvis, 2007). The presence of preferential transport reduces the contact time of solutes with the soil matrix, making their adsorption difficult and resulting in significant environmental issues, such as groundwater pollution. Additionally, preferential flow influences the hydrological cycle, soil erosion, agricultural production, and various biochemical and ecological processes within the vadose zone. It has a positive influence over flood regulation, erosion prevention, and aquifer recharge, but a negative impact in terms of faster contaminant transport, thus affecting water quality (Guo and Lin, 2018).

The effects of vegetation are two-sided. On the one hand, the root system increases the abundance of soil pores. On the other hand, the presence of the vegetation cover has resulted in a decrease of preferential flow (Stumpf and Maloszewski, 2010). Soil fauna has the same positive influence as the vegetation. Generally, soil fauna influences many physical and chemical processes in soil (Kooistra and Pulleman, 2018). It has an essential role in altering various soil processes, such as organic matter decomposition and soil structure formation. Soil compaction from trafficking, which frequently occurs in intensively managed arable landscapes, can increase lateral preferential flow and generate the destruction of the macropore system (Jarvis, 2007; Sander and Gerke, 2007).

To decrease nutrient and pesticide losses, and thus to protect surface and subsurface water resources, extensive understanding of soil water preferential flow and transport of dissolved substances is fundamental (Salvador et al., 2011). Nonequilibrium flow and transport in structured soil are highly complex. The characterization of these processes thus requires a combination of various methods, which can result in their better quantification.

Dye staining is an efficient method for assessing preferential flow in soils at different scales (e.g., column, profile). Dyes with the following characteristics are used for the monitoring of water flow within the soil profile and the estimation of soil pore system appearance, size, and distribution (Flury and Flühler, 1994, 1995): visibility, non-toxicity, non-accumulation in soil flora and fauna, and minimal subsequent degradation in the soil. The most commonly used dye tracer in soil hydrology on both field and column scale is Brilliant Blue FCF (e.g., Fuentes et al., 2014; Schneider et al., 2018). For example, Bejbe et al. (2017) described the interactions between the Brilliant Blue dye solution with soil particles and their effect on the mobility of substances around the zones of preferential flow. Sorption of Brilliant Blue FCF in soils was found to be affected by soil pH, ionic strength, and soil composition (Germán-Heins and Flury 2000). Kodešová et al. (2015) confirmed that dye distribution is significantly affected by the presence of large pores due to the earthworm population, root system spreading, and organic matter in the soil. It was also noted that the influence of topology, macropores, and soil water content is important.

However, dye staining is a destructive method when used on soil samples. A dye can sorb to macropore walls, resulting in an overestimation of nonequilibrium flow patterns. Thus, an advanced non-invasive and non-destructive X-ray computed tomography (CT) method is often used when precise quantification of the soil pore system needs to be achieved. It is a method superior to dyeing because of a high spatial resolution, which clearly exposes and differentiates soil pores, roots, and solid-phase fractions. In soil science, the CT method is applied for monitoring of time-dependent soil physical properties such as porosity, pore characteristics, density, meandering, and volumetric water content, as well as solute transport parameters, fractal characteristics, soil aggregation, and retention and hydraulic conductivity curves under unsaturated conditions (Cercioglu, 2018; Périard et al., 2016; Rogasik et al., 2003; Tracy et al., 2015). The method has considerable potential for the quantification of different processes within the soil profile, such as root development, soil structure, water flow, and solute transport (Koestel, 2017). However, the implementation of X-ray computed tomography alone is often not sufficient to study non-linear processes (i.e., flow and transport) in heterogeneous arable soils.

Numerical models have been developed to describe such nonequilibrium flow processes in structured soils. Commonly used, physically-based numerical models for simulating water flow and solute transport in soils use either single continuum or bi- and multi-continuum approaches (Kodešová et al., 2010). Bi-modal concepts for nonequilibrium flow assume that the porous soil system is divided into two domains, in which one domain explains matrix (micropore) flow and the other fracture (macropore) flow, with two sets of transport properties (Gerke and van Genuchten, 1993). Haghverdi et al. (2020) studied the performance of five unimodal water retention models and eleven combinations of alternative (e.g., Peters–Durner–Iden PDI) and bi-modal expressions on 94 soil samples with high-resolution data sets from Turkey and the United States. Overall, Haghverdi et al. (2020) showed that alternative expressions provided a better fit than unimodal expressions. To correctly characterize the soil and to parameterize more complex models, extensive and reliable data sets need to be analyzed by various techniques. Guo et al. (2019) combined time-lapse ground-penetrating radar, controlled infiltration, and high-frequency moisture monitoring with a 2D/3D numerical modeling to investigate preferential flow in saprock. As concluded by Guo et al. (2020), the pairing of a useful combination of geophysical methods is needed to advance our understanding of the complex interactions between plant (e.g., tree roots) and soil. Guo and Lin (2018) suggested that the magnitude and spatial pattern of preferential flow are regulated by interactions among soil architecture, soil properties, landscape setting, and land use/land cover, which change with seasonal soil wetness conditions and water input characteristics.

The overall goal of this study was to assess the soil porous system and preferential flow pathways in vineyard soil by combining laboratory and numerical methods with dye staining and X-ray imaging. It was hypothesized that the integration of various methods could reveal more information regarding soil structure and flow behavior in structured arable soil. The specific objectives were i) to identify soil hydraulic parameters for two conceptual models (i.e., single-porosity and dual-permeability models) by combining laboratory (evaporation and leaching experiments) and numerical approaches, ii) to visualize dye staining (Brilliant blue, BB) patterns within the vertical soil column sections, (iii) to estimate and visualize pore architecture using X-ray tomography, iv) to compare the BB stained soil images and X-ray images to distinguish flow in macropores and matrix pores, and v) to approximate dye transport using the two conceptual models (single-porosity and dual-permeability) and thus to identify the more suitable model.

2. Materials and methods

2.1. Study site and soil column preparation

The soil column study was performed in the laboratory of the Department of Soil Amelioration (MELILAB), University of Zagreb, conventional tillage; Cerdan et al., 2010). Thus, vineyard soils present an interesting research topic about soil structure heterogeneity and possible occurrence of preferential flow.
Faculty of Agriculture (UNIZG-AGR), using soil samples from the Experimental Station Baštica (44°09′19″N, 15°26′34″E), located near Zadar, Croatia, and managed by the Department of Viticulture and Enology, UNIZG-AGR. The vineyard site occupies 6.5 ha and has a long-term average annual temperature of 15.5 °C and rainfall of 879 mm. The vineyard was planted in 2008 with native Dalmatian varieties of V. vinifera L. cv. Pošip and Marastina. For the determination of soil textural composition and physicochemical properties, disturbed soil sampling was performed (at 0–25, 25–50, 50–75, and 75–125 cm soil depths) in triplicates (Table 1). Undisturbed soil columns were taken within vineyard plant lines by gently pushing the PVC tubes (n = 7, 40 cm long with a 16 cm diameter) into the soil to minimize the disturbance of micro and macroporosity. Before the sampling, in order to prevent the edge effect of water leaching alongside column walls, a non-reactive glue was applied to the column walls (e.g., Kodešová et al., 2010). One column was used for X-ray scanning, while the remaining six columns were used for dye staining, three of which were subsequently used for dye staining. The soil pH was measured using the Mettler Toledo MPP 227 conductivity/ph-water in meter (pH H₂O) (HRN ISO 10390:2005). The organic matter content was determined by sulfochromic oxidation (HRN ISO 14235:1998). The soil particle size distribution (fractions of sand, silt, and clay) was determined using the pipette method (Gee and Or, 2002). The soil was classified according to the IUSS (2014) as Calcaric Leptosol. Soil pH, texture, bulk density, and organic matter content are presented in Table 1.

### 2.2. Soil hydraulic parameters and water flow estimation

Undisturbed soil cores (250 cm³, n = 5) were taken at multiple depths of the topsoil horizon, up to 40 cm (5–10 cm, 10–15 cm, 20–25 cm, 30–35 cm, and 35–40 cm). The saturated hydraulic conductivity $K_s$ [LT⁻¹], was determined using the KSAT instrument (METER Group, USA) on these soil cores. The working principle of the KSAT instrument is based on Darcy’s equation. After $K_s$ estimation, the remaining soil hydraulic parameters (SHP) were estimated on the same undisturbed soil cores (250 cm³) using the simplified evaporation method (Schindler and Müller, 2017) and the HYPROP automatized system applicable to the most soil types (Haghighi et al., 2018). This method takes into account the change of the sample weight and a matrix potential in the soil sample during the evaporation drying process (METER Group, USA). Soil hydraulic properties (SHP) for Calcaric Leptosol soil columns were determined using the HYPROP-FIT program and are shown in the Results section (Table 2).

SHPs were described using the van Genuchten-Mualem model (VGM, van Genuchten, 1980):

\[
\theta(h) = \theta_s - \theta_r \left( \frac{h}{L} \right)^m \quad \text{for } h < 0
\]

\[
\theta(h) = \theta_s + \frac{\theta_r - \theta_s}{1 + \left( \frac{h}{L} \right)^n} \quad \text{for } h \geq 0
\]

\[
K(h) = K_s \left( 1 - (1 - \theta_s)^{2n} \right)^{1/2}
\]

where $\theta_s$ and $\theta_r$ denote residual and saturated volumetric water contents [L³ L⁻³], respectively, $h$ is the pressure head [L], $S_e$ is the effective saturation [–], $\alpha$ [L⁻¹] and $\theta$ [–] are shape parameters, and $m$ [–] is a pore connectivity parameter. Pore connectivity parameter, $t$, is fixed in this study to a value of 0.5 (Mualem, 1976).

Undisturbed soil columns were saturated and left to drain in order to reach the steady-state (matching) conditions before the intermittent leaching experiment (n = 6). The bottom of the soil columns was covered with a fine plastic mesh and tightly taped to prevent any soil material disturbance. The experiment was conducted in seven cycles carried out over 23 days. Approximately 350 mL of irrigation water was applied during each application (divided to 50 mL doses that were applied over a few hours to prevent excessive waterlogging) at the top of the columns in the form of mist using a hand-help sprayer to ensure uniform water distribution. Water samples (leachate) were collected at the bottom of the column after each application and volumes measured.

#### 2.3. Numerical modeling

Numerical modeling was performed using the HYDRUS-1D program (version 4.0) (Šimůnek et al., 2016) that uses the Galerkin-type linear finite element scheme to solve the governing flow equations for single-porosity (SP) and dual-permeability (DP) models. Water flow simulations (of intermittent leaching experiments) were based on a numerical solution of the Richards equation for variably-saturated water flow in a single-porosity (SP) media representing soil columns:

\[
\frac{\partial \theta}{\partial t} = \frac{\partial}{\partial z} \left( K(h) \frac{\partial \theta}{\partial z} \right) + q_c
\]

where $\theta$ is the volumetric water content [L³ L⁻³], $h$ is the water pressure head [L], $K$ is the hydraulic conductivity [LT⁻¹], $t$ is time [T], and $z$ is the vertical coordinate (positive upward) [L].

The advection–dispersion equation was used to simulate the solute transport (Brilliant blue dye):

\[
\frac{\partial c}{\partial t} = \frac{\partial}{\partial z} \left( D \frac{\partial c}{\partial z} - qc \right)
\]

where $c$ is the solute concentration in the liquid phase [M L⁻³], $q$ represents the volumetric water flux density [LT⁻¹] and $D$ is the dispersion coefficient [L²T⁻¹] defined as follows:

#### Table 1

<table>
<thead>
<tr>
<th>Depth [cm]</th>
<th>Organic matter [%]</th>
<th>Soil texture [%]</th>
<th>pH_H₂O</th>
<th>EC [dS m⁻¹]</th>
</tr>
</thead>
<tbody>
<tr>
<td>0–25</td>
<td>1.14</td>
<td>71</td>
<td>19</td>
<td>10</td>
</tr>
<tr>
<td>25–50</td>
<td>1.00</td>
<td>70</td>
<td>17</td>
<td>13</td>
</tr>
<tr>
<td>50–75</td>
<td>0.76</td>
<td>65</td>
<td>19</td>
<td>16</td>
</tr>
<tr>
<td>75–125</td>
<td>0.69</td>
<td>63</td>
<td>18</td>
<td>20</td>
</tr>
</tbody>
</table>

* EC electrical conductivity.

#### Table 2

<table>
<thead>
<tr>
<th>Sample No.</th>
<th>Depth [cm]</th>
<th>Porosity [%]</th>
<th>$\theta_c$ [L³ L⁻³]</th>
<th>$\theta_a$ [L³ L⁻³]</th>
<th>$a_c$ [cm⁻¹]</th>
<th>$n$ [–]</th>
<th>$K_s$ [cm d⁻¹]</th>
<th>RMSE_TH [cm³ cm⁻³]</th>
<th>RMSE_K [cm d⁻¹]</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>5–10</td>
<td>43</td>
<td>0.031</td>
<td>0.395</td>
<td>0.019</td>
<td>1.56</td>
<td>3.00</td>
<td>0.017</td>
<td>0.82</td>
</tr>
<tr>
<td>2</td>
<td>10–15</td>
<td>42</td>
<td>0.087</td>
<td>0.371</td>
<td>0.020</td>
<td>1.50</td>
<td>4.20</td>
<td>0.002</td>
<td>0.16</td>
</tr>
<tr>
<td>3</td>
<td>20–25</td>
<td>40</td>
<td>0.070</td>
<td>0.361</td>
<td>0.029</td>
<td>1.53</td>
<td>12.80</td>
<td>0.006</td>
<td>0.25</td>
</tr>
<tr>
<td>4</td>
<td>30–35</td>
<td>40</td>
<td>0.088</td>
<td>0.380</td>
<td>0.018</td>
<td>2.00</td>
<td>2.90</td>
<td>0.009</td>
<td>0.39</td>
</tr>
<tr>
<td>5</td>
<td>35–40</td>
<td>37</td>
<td>0.057</td>
<td>0.358</td>
<td>0.016</td>
<td>1.61</td>
<td>3.86</td>
<td>0.01</td>
<td>0.39</td>
</tr>
</tbody>
</table>
\[ \theta D = D_{t} |q| + \theta D_{w} \tau \]  
(7)

where \( \theta D_{t} \) is the coefficient of molecular diffusion [ML^{2}T^{-1}], \( \tau \) is the tortuosity factor [~], and \( D_{t} \) represents the longitudinal dispersivity [L].

Additional modeling of intermittent leaching experiments was also performed using a dual-permeability model, in which the Richards equation is applied separately to two overlapping pore regions, i.e., the macropore and matrix domains (Gerke and van Genuchten, 1993). A set of two coupled Richards equations for flow in the fracture and matrix domains, respectively, is defined as follows:

\[ \frac{\partial \theta_{f}(h_{f})}{\partial t} = \frac{\partial}{\partial z} \left\{ \frac{K_{f}(h_{f})}{\partial z} + 1 \right\} - \frac{\Gamma_{w}}{f_{w}} \]  
(8)

\[ \frac{\partial \theta_{m}(h_{m})}{\partial t} = \frac{\partial}{\partial z} \left\{ \frac{K_{m}(h_{m})}{\partial z} + 1 \right\} + \frac{\Gamma_{w}}{1 - f_{w}} \]  
(9)

where the subscripts \( f \) and \( m \) refer to fracture (macropore) and matrix (micropore) domains, respectively, \( \Gamma_{w} \) is the water transfer term for water exchange between the macropore and matrix domains [T^{-1}], and \( f_{w} \) and \( f_{w} \) is the macropore domain fraction [-]. The matrix domain fraction is specified as \( 1 - f_{w} \). The total composite soil water content and hydraulic conductivity are defined as the sum of the soil water contents and hydraulic conductivities of each domain multiplied by corresponding domain fractions.

The mass exchange between the matrix and macropore regions, \( \Gamma_{w} \), is expressed as:

\[ \Gamma_{w} = \frac{\beta}{a^{2}} \kappa_{e}(h_{f}) \pi_{w} (h_{f} - h_{m}) \]  
(10)

in which \( \pi_{w} (0.4) \) is a dimensionless scaling factor (Gerke and van Genuchten, 1996), and \( K_{e} \) is the effective hydraulic conductivity of the interface between the two pore domains [L T^{-1}], defined as:

\[ K_{e}(h_{f}) = K_{o}m(h_{f}) \]  
(11)

where \( K_{o}m \) is the saturated hydraulic conductivity of the interface between the macropore and soil matrix domains, \( K_{o}m(h_{f}) \) is the relative unsaturated hydraulic conductivity of the matrix domain calculated using the van Genuchten model for the pressure head of the interface \( h_{o} \) (taken as \( h_{f} + h_{m}/2 \)). The dimensionless shape factor \( \beta \) (15 for spherical aggregates, 3 for cubic aggregates), and the characteristic length of an aggregate \( a \) (a sphere radius or half size of the cube edge) are the parameters describing the aggregate shapes (Gerke and van Genuchten, 1996).

The dual-permeability formulation for solute transport is based on two coupled advection–dispersion equations (Gerke and van Genuchten, 1993):

\[ \frac{\partial c_{f}}{\partial t} = \frac{\partial}{\partial z} \left\{ \beta_{f} D_{f} \frac{\partial c_{f}}{\partial z} \right\} - \frac{\partial \theta_{f} c_{f}}{\partial z} - \frac{\Gamma_{w}}{f_{w}} \]  
(12)

\[ \frac{\partial c_{m}}{\partial t} = \frac{\partial}{\partial z} \left\{ \beta_{m} D_{m} \frac{\partial c_{m}}{\partial z} \right\} - \frac{\partial \theta_{m} c_{m}}{\partial z} + \frac{\Gamma_{w}}{1 - f_{w}} \]  
(13)

where \( \Gamma_{w} \) is the solute mass transfer term [M L^{-3} T^{-1}] (Gerke and van Genuchten, 1996):

\[ \Gamma_{w} = \alpha_{w}(1 - f_{w}) \theta_{w} (c - c_{w}) + \Gamma_{w} c^{*} \]  
(14)

where \( c^{*} \) is either \( c_{f} \) or \( c_{m} \) depending on the exchange direction, and \( \alpha_{w} \) is the first-order solute mass transfer rate coefficient [T^{-1}] of the form:

\[ \alpha_{w} = \frac{\beta}{a^{2}} D_{w} \]  
(15)

in which \( D_{w} \) is an effective diffusion coefficient [L^{2}T^{-1}], which represents the diffusion properties of the fracture-matrix interface (e.g., accounting for organic coatings).

The parameters for the single-porosity model were derived from the evaporation experiment. After a single-porosity model run, the dual-permeability model was applied to capture preferential flow leaching. The fraction of the macropore domain \( f_{w} \) was determined based on the average pore distribution (macropores) derived from the CT images of the scanned soil column (see the Results and Discussion Section below). Parameters characterizing the aggregate geometry were set following the study of Kodešová et al. (2010). The following parameters were used to define the structure of both domains: \( \beta = 3 \) (the shape factor characterizing cubic aggregates), \( a = 2 \text{ cm} \) (the characteristic length of an aggregate defining elongated pathways), and \( K_{o}m = 1.0 \times 10^{6} \text{ cm h}^{-1} \). The effective saturated hydraulic conductivity of the macropore-matrix interface was set relatively low to avoid any numerical instabilities (Kodešová et al., 2010). The retention curve parameters \( \theta_{s}, \theta_{r}, \alpha, \) and \( n \) were used to characterize matrix (subscript \( m \)) and macropore (subscript \( f \)) domains where saturated volumetric water contents were kept the same for both domains while residual water contents were set to 0 (Gerke and Kohne, 2004; Kodešová et al., 2008, 2010). The parameters \( \theta_{s} \) and \( \theta_{r} \) were increased to 0.1 cm^{-1} and 3 for the entire column domain, respectively. It has been found previously that such values increase macropore flow (Kodešová et al., 2010). While the saturated hydraulic conductivity of the matrix domain, \( K_{o}m \), was determined using the evaporation experiment (Table 2), the saturated hydraulic conductivity of the macropore domain, \( K_{o}f \), was increased by 100 to account for macropore flow. A related \( K_{o}f/K_{o}m \) ratio was estimated in Kodešová et al. (2008). Solute mass transfer coefficient, \( \alpha_{w} \), was set to 0.02 d^{-1} (Gerke et al., 2013). It should be noted here that the objective of this work was not an in-depth analysis of the dual-permeability model parametrization.

Initial conditions for modeling were set to the pressure head of 0 at the bottom and −40 cm at the top (hydrostatic equilibrium) of the soil column. A time-variable atmospheric flux boundary condition (applied irrigation and evaporation) was used at the top, and a seepage face boundary condition was applied at the bottom of the column (re-plicating experimental procedure in Section 2.2). Dye simulation followed the same initial and boundary condition with additionally adding solute transport and adapting input dosage and amount as explained in Section 2.4.

1 – \( f_{w} \). The performance of the calibrated water flow model was evaluated using the root mean square error (RMSE, for evaporation and leaching experiments) and the coefficient of determination (\( R^{2} \), for leaching experiments). The model error for water retention (RMSE, TH) was calculated separately from the model error for hydraulic conductivity (RMSE, K):

\[ \text{RMSE} = \sqrt{\frac{\sum_{i=1}^{N} (O_i - P_i)^2}{N}} \]  
(16)

\[ R^{2} = \frac{\sum_{i=1}^{N} (O_i - \bar{O})(P_i - \bar{P})}{\left[ \sum_{i=1}^{N} (O_i - \bar{O})^2 \left( \sum_{i=1}^{N} (P_i - \bar{P})^2 \right)^{1/2} \right]} \]  
(17)

where \( O_{i} \) are observation data points, \( P_{i} \) are model predictions, \( \bar{O} \) is an average observation, \( \bar{P} \) is an average prediction, and \( N \) is the sample size.

2.4. Dye staining experiments and image analysis

The dye experiments with soil columns were performed under controlled conditions in MELILAB at the UNIZG-AGR after the intermittent leaching experiment (Fig. 1). Brilliant blue dye (Brilliant Blue FCF) in the form of solute (5 g L^{-1}) was added to the soil columns (n = 3). The dye solution was dissolved in 500 mL of distilled water and applied (divided into 250 mL dosages) with a handheld sprayer so that approximately 1 cm of ponding occurred at the column top. After 24 h, columns were cut along the longer axis (vertically) in half. From each column side, 2 cm slices (40 cm long) were prepared, resulting
altogether to eight slices per column. Soil slices were then photographed using a high-resolution camera (Samsung NX1000, 20.3 MP; 20–50 mm f/3.5–5.6 ED II Lens). Image analysis was performed using the ImageJ software, commonly used for image processing and analysis (Ferreira and Rasband, 2012). The images were “thresholded” after correction to create binary images with stained and unstained areas. Dye coverage was calculated from the percentage of stained pixels vs. depth, according to Flury et al. (1994) and Sander et al. (2007). ImageJ was used for the determination of soil stained by Brilliant blue dye and for the quantification of the pore volume and distribution inside the soil column. In these analyses, only the upper 30 cm of soil samples were taken into account to avoid disturbed soil at the bottom of the column. Also, the applied Brilliant Blue FCF dye colored the soil down to the 30 cm depth.

2.5. X-ray imaging of soil columns

The selected soil column was scanned using X-ray computed tomography (CT) Siemens Somatom Definition AS to visualize and quantify internal soil porosity. The column was scanned using a medical-grade scanner at a voltage of 120 kV and a current of 350 mAs. Resolution was 0.48 mm × 0.48 mm × 0.48 mm. Only the upper 600 radiographs were selected for the analysis as the soil column showed signs of disturbance in the bottom part (likely occurring during transport and handling). Pictures recorded for the bottom of the soil column were therefore discarded as the soil started to dissipate (due to the higher sand content in that layer, Table 1). By conducting the image analysis, the pore volume and distribution were determined, as well as the 3D representation of the porous soil system. The radiographs were subsequently inverted into 3D images and exported as TIFF-stacks (a tagged image file format). Image processing of the X-ray and dye experiments was accomplished using the open-source software ImageJ and the bundle of plugins distributed by FIJI (Schindelin et al., 2012). Since all X-ray parameters and settings were kept the same for all images, image reconstruction of all layers could be accomplished on a semi-automated basis. Fixed threshold values, to distinguish between pores and the solid phase, were selected based on the method presented in the SoilJ plugin (Koestel, 2017). Two gray reference values were selected that correspond to objects of known or at least constant density. Typically, one of the reference values corresponds to the gray value of the column walls. The second gray value was chosen as a quantile of the histogram of gray-values within individual horizontal cross-sections. A very low quantile, e.g., 0.001, was chosen to represent the least dense phase in the imaged object (i.e., air). X-ray scanning and image analyses were performed using the methodology presented by Larsbo et al. (2014). Image processing (thresholding and segmentation) and analysis were carried out using the Fiji distribution (Schindelin et al., 2012) of the ImageJ software (Abramoff et al., 2004), including the BoneJ (Doube et al., 2010) and SoilJ (Koestel, 2017) plugins. The main output of X-ray CT scanning was the visualization of the 3D pore system architecture, the quantification of the pore system, and the estimation of the pore origin, e.g., bio pores or cracks.

3. Results and discussion

3.1. Soil hydraulic parameters and water flow

The SHPs of the VGM model are shown in Table 2. Errors in the water retention and hydraulic conductivity model were below 0.02 cm³ cm⁻³ and 0.6 cm day⁻¹, respectively, indicating that the unimodal van Genuchten (VGM) model was appropriate for representing the soil hydraulic properties during the evaporation experiment. Table 2 shows that the saturated hydraulic conductivity decreased in the top and bottom of the soil column, where values were 3.0 and 3.86 cm day⁻¹, respectively. The highest hydraulic conductivity was at a depth of 20–25 cm, where it was 12.8 cm day⁻¹. The soil surface appears to be affected by heavy trafficking in the vineyard, and the absence of roots and macrofauna. The middle layer had higher porosity, resulting in higher hydraulic conductivity. It is interesting that the bi-modal characteristic of the capillary pore size distribution, usually present in structured soils (Kodešová et al., 2009, 2012), was not detected here using the classical approach (Fig. 2). Also, one layer (i.e., the first 5–10 cm layer) did not follow the VGM fit well. Indeed, when the bi-modal VGM model was used in this soil layer, the improved fit was observed (RMSE,TH = 0.0035 cm³ cm⁻³ and RMSE,K = 0.3143 cm day⁻¹). However, such improvements were not found in other soil layers, and thus the single-porosity VGM model was applied.

As reported in Bezerra-Coelho et al. (2018), most soil types ranging from fine to very coarse textures can be described using the single-porosity VGM model. The dual-porosity model (Durner 1994) is applicable only in cases when a good fit using a single-porosity VGM model cannot be obtained. When a dual-porosity model of Durner (Durner 1994) is used, an apparent double S-shape curve in the soil water retention curve is expected to fit the experimental data. It should be noted here that the bi-modal character of the pore structure can be hidden (Kodešová et al., 2009, 2008) even when the pore system is indeed bi-modal and needs to be divided into two regions (Gerke and Kohn, 2004; Gerke and van Genuchten, 1996; Kodešová et al., 2010; Vogel et al., 2000). In the Introduction, we cite Haghverdi et al. (2020), who reported for a large dataset that the bi-modal and alternative expressions (e.g., including Peters-Durner-Iden PDI) could provide a better fit than unimodal models. Still, the differences are sometimes almost negligible (e.g., VG: RMSE = 0.015, r (correlation) = 0.995 vs. VG-PDI RMSE = 0.009 r = 0.998; Haghverdi et al., 2020). Thus, in such situations, both hydraulic property models would provide a very similar description of flow behavior. This issue is further explored below.

Additionally, the calculated porosity on the column scale was 44.2% (n = 6), showing comparable results to those obtained using HYPROP (40.4%, n = 5). A decrease in soil porosity and an increase in the clay content of the last observed soil column part (Table 2) were in line with decreasing hydraulic conductivity.
Although the single-porosity VGM model provided an excellent overall description of water flow (Fig. 3a, R² of 0.984), the dye staining and X-ray imaging presented evidence of extended macropore network (see Chapters 3.2.1 and 3.2.2), which should result in preferential flow for conditions close to saturation. Thus, as reported previously (Kodešová et al., 2006; Kodešová et al., 2008; Filipović et al., 2019), a different modeling approach, i.e., a dual-permeability model, can differentiate between flows in soil macropore and matrix regions. Additional dual-permeability modeling was thus performed using HYDRUS-1D. Similar results for the intermittent leaching experiments (Fig. 3) were obtained using the SP and DP models in terms of goodness of fit (R² SP = 0.984; R² DP = 0.987). Both modeling approaches, SP and DP, showed a similar water flow behavior and resulted in a good fit when compared to the experimental data involving cumulative outflow. A comparable observation was found by Kodešová et al. (2008), who used the SP and DP models for multistep outflow data collected on Greyic Phaeozem soil. In our leaching experiments, the use of the DP model, compared to the SP model, resulted in a faster response of outflow to irrigation and higher outflow rate peaks. Unfortunately, the experimental data collected during the performed leaching experiments did not provide sufficient evidence to distinguish between the two models and between equilibrium and nonequilibrium flow patterns. While one would not expect to obtain such evidence from evaporation experiments, which is run under unsaturated conditions, such evidence could have been obtained from leaching experiments, had more detailed outflow dynamics been observed (more, rather than only two, data points for each leaching cycle), or had environmental tracers been used in infiltrating water (observing outflow breakthrough curves with arrival times, peaks, and possible tailing).

It is also well known that the soil hydraulic properties estimated using different methods can be quite different even in the case of sandy soil, known for its uniformity (e.g., Gribb et al., 2004). Thus, indicating
the necessity of implementing various methods and experiments during transient flow and transport studies in arable soils. Besides the effect of multimodality of the soil porous systems on soil water regimes, another reason may be the hysteresis in soil hydraulic functions. Other factors affecting resulting soil hydraulic properties may include the character of the experiment (i.e., steady-state or transient flow experiments), the size of the soil sample, dimensions of the flow domain (1D, 2D, or 3D), and methods used for evaluation of soil hydraulic properties (analytical equations or inverse modeling). Kutilek et al. (2009) tested empirical, semi-empirical, and physically-based models of soil hydraulic functions derived for bi-modal soils aimed at capturing preferential flow. They concluded that, although many empirical models exist that could be modified for multimodal functions, simple models often provide a good agreement between modeled and measured flow data. This implies that numerical models simulating water flow using unimodal models can still provide a good agreement with experimental data on water flow collected in structured soils. However, such models then often fail to provide a good description of the solute transport behavior.

3.2. Identification of flow in preferential pathways and matrix pores

3.2.1. Soil column dye staining

Dye staining patterns in vertical cross-sections of the soil columns indicated the highly heterogeneous nature of the flow patterns in this arable soil. Percentages of stained areas for column 1 are in the range from 17.2% to 33.4%, for column 2 from 18.5% to 31.9%, and for column 3 from 18.4% to 25.3% (Table 3). Figs. 4 to 6 illustrate the uneven dye distribution caused by the enhanced dye penetration along biopores and in regions with a higher fraction of larger interaggregate pores formed in the root zone. A significant preferential flow could be observed below the soil depth of approximately 10 cm, except in Fig. 4, sections 1 – 4b, where preferential flow started to occur below 5 cm. This was also confirmed when analyzing SHPs since the middle layer (20–25 cm) had higher hydraulic conductivity (Table 2). Similar observations were reported on a field scale by Kodešová et al. (2012). They found the occurrence of extensive preferential flow in the subsurface horizons of arable soils, i.e., Hapluc Luvisol and Hapluc Cambisol.

In our study, the plow pan effect was observed below the depth of 30 cm, which is probably due to the vineyard tillage management, which is usually limited to the approximately upper 20–30 cm soil depth between the rows (e.g., Comino et al., 2017). This is illustrated in dye staining images and supported by lower porosities in deeper soil layers. Sander and Gerke (2007) reported that dye penetration in drained paddy soils was directed horizontally when plow pan was reached, while further penetration to deeper layers occurred through biopores present in the plow pan.

By additionally estimating dye staining soil patterns, information regarding macropores origin could help in clarifying the preferential flow behavior. This is further explored in the next section, where the dye staining and X-ray imaging results are compared. The aggregate coatings also play an important role in dye transport and visualization. Pore geometry in the soil changes with depth, thus influencing the spatial orientation of the preferential flow (Guo and Lin, 2018). Kodešová et al. (2015) found that dye distribution improved when straw residues, earthworms, and roots were present in the soil, but also that topology, macropores, and water content have an impact on dye distribution as well. Presented dye staining images indicate that the main part of the macropore network is formed by root channels and earthworm burrows. Although macropore flow can be determined using this method, limitations exist due to potential dye sorption onto soil particles. Nevertheless, preferential flow and infiltration patterns in the vadose zone can be reliably determined by the application of Brilliant Blue dye (Wu et al., 2015).

3.2.2. X-ray Imaging, pore architecture, and bimodal system conceptualization

From 600 images produced using the X-ray CT scanning and processed using the Image J software, image No. 8 had the largest fraction of the soil matrix within the sample column. In contrast, images No. 139 and 140 had the smallest fraction (Fig. 7). There were twenty column sections with average values of 6.6% of macropores. Despite equal percentages of the soil matrix in images No. 87 and 568, it is visible that the pore arrangement in these column sections is entirely different. The difference in the pore arrangement is caused by various factors, mostly by the presence of flora and fauna in the soil (e.g., Menta, 2012). Images 1, 200, 400, and 600 (Fig. 8a) are chosen to illustrate changes in the appearance and distribution of pores at different soil depths. The fractions of the macropores within the selected column are in the range from 2.3 to 1%. Additionally, an unprocessed image from X-ray scanning is shown in Fig. 8b to illustrate the scale of the macropore network. The largest pores had diameters from 5.2 up to 10.6 mm, suggesting that indeed the pore origin is from earthworm burrows and roots. The preferential flow through the large diameter macropores is well known to occur in arable soils. These macropores are usually described as biopores (e.g., Beven and Germann, 2013; Guo and Lin, 2018; Sander and Gerke, 2007).

If we compare the results obtained by dye staining and CT, i.e., the average percentages of the pore systems identified by the two methods (23% compared to 7%, respectively), there is a large difference of 16%. This difference can be related to dye coloring also the soil around macropores, i.e., it can be related to dye penetration into the pores smaller than the CT threshold. Again, this illustrates the difficulty of selecting an appropriate method to quantify preferential flow in structured soils. Soto-Gómez et al. (2019) presented a novel method of combining fluorescence macrophotography and X-ray computed tomography (CT) in preferential flow estimation. They concluded that preferential flow occurred in only a small fraction of the total pore network and was controlled by pores connected to the soil surface and by matrix density. This can be further illustrated by overlapping the dye staining and X-ray imaging. Fig. 9a shows the macroporosity detected using the X-ray scanning and the percentage of soil stained with Brilliant Blue FCF dye. The latter shows that dye was transported from the}

<table>
<thead>
<tr>
<th>Column No.</th>
<th>Image No.</th>
<th>Stained area [%]</th>
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</thead>
<tbody>
<tr>
<td>Column 1</td>
<td>1a</td>
<td>26.7</td>
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<tr>
<td></td>
<td>2a</td>
<td>31.1</td>
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<td></td>
<td>1b</td>
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<td></td>
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<td></td>
<td>3b</td>
<td>19.4</td>
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<tr>
<td></td>
<td>4b</td>
<td>18.2</td>
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<tr>
<td>Column 2</td>
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<tr>
<td></td>
<td>2a</td>
<td>23.2</td>
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<td></td>
<td>3a</td>
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<tr>
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column surface down to a depth of about 8–10 cm in the soil matrix pores, while the deeper presence of dye can be explained only by the transport in macropores. Numerical modeling of the Brilliant Blue transport (Fig. 9b) with HYDRUS-1D indicate that a single-porosity model cannot explain dye coloring in larger depths (i.e., below 15 cm) while the dual-permeability model can capture the preferential flow in the macropores as well (i.e., in entire soil column). It should be noted here that Fig. 9a represents dye coloring which is considerable lighter at the 10–20 cm depth (Figs. 4 to 6). Contrary Fig. 9b represents Brilliant Blue mass which should explain the discrepancy between the two.

In some instances, according to Nielsen (2004), porosity determined by X-ray CT differs from values obtained in the laboratory. However, X-ray CT scans can be used to identify the largest pores present in the system and for macropore detection. By decreasing the image quality, the accuracy of the porosity determination is reduced as well. This is well known, especially when relatively large soil samples are used, as was done in our study. A similar conclusion was made by Rab et al. (2014). They concluded that sample scanning at a higher resolution would result in more macropores detected, compared with sample scanning at a lower resolution. One of the possible error origins is the
determination of thresholds between the solid soil phase and pores during image processing, due to the subjective perception involved in this step. However, in our study, the aim was to capture the pore architecture on a column scale, thus accepting the lower resolution of X-ray images. Fig. 10 displays a three-dimensional (3D) pore system architecture obtained using X-ray on a column taken from the Calcaric Leptosol soil. From the 3D visualization, it is clear that biopores originating from earthworm burrows or roots represent most of the pore...

Fig. 6. Images of soil after the application of the brilliant blue dye (Brilliant Blue FCF) (left image with brown background) and the ImageJ processing (right image) for soil Column 3 cut in half (marked as a and b) and then sliced to 4 column slices (1–4). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Fig. 7. Column X-ray images with the maximum (image No. 8 at 3.84 cm depth), minimum (image No. 139 at 6.67 cm depth), and average (images No. 87 at 4.18 cm depth and 568 at 27.26 cm depth) solid phase (soil matrix) percentages. The soil pore system is represented in white color.
Leue et al. (2019) reported that the quantitative techniques, such as, e.g., X-ray CT combined with subsequent (3D) image analysis, are needed to differentiate between biopores and cracks. Here, a more holistic approach was used, in which a combination of methods resulted not only in an overall description of the pore system but also of the flow behavior. A relatively large column size (40 cm long × 16 cm in diameter) did not allow for much better image resolution using the medical scanner. However, by combining all presented methods, the preferential flow was quantified, and its origin determined. Gao and Lin (2018) reported that 25% of 190 analyzed case studies on preferential flow combined multiple methods for its quantification. As also proven in this study, the integration of different methods overcomes some of the limitations of each technique and helps to constrain the detected preferential flow patterns and reveals some hidden information.

4. Conclusions

This study combined laboratory flow experiments and numerical methods with dye staining and X-ray imaging to assess pore system architecture and preferential flow in the vineyard soil. Soil hydraulic properties measured in the laboratory and also determined using numerical modeling did not provide enough evidence supporting the nonequilibrium flow patterns. The numerical simulations performed well when the single-porosity model was used to simulate leaching experiments. However, because the extensive macropore network was...
revealed using dye staining and X-ray imaging, the dual-permeability model was used as well. The dual-permeability model also produced a high level of agreement ($R^2 = 0.987$), while additionally providing a faster response to applied irrigation events. However, collected experimental data did not allow us to differentiate the performance of the two models. The study showed that the bi-modal character of pore structure could often be hidden when a limited number of methods or non-adequate methods (e.g., by relying only on laboratory-derived SHPs and numerical modeling) are applied for its quantification.

The dye patterns, which represented about 17 to 33% (average 23%) of the area within the vertical column sections, indicated the presence of preferential (macropore) flow in the arable Calcaric Leptosol vineyard soil. Additionally, a combination of dye staining and X-ray imaging revealed a complex pore architecture network. Using only dye staining can also be misleading due to dye sorption properties or relatively large mass exchange in preferential flow quantification studies. The preferential flow was mostly caused by biopores, i.e., earthworm burrows and root channels (7% of the soil column). The macropores with a diameter of up to 10.6 mm were found during the image analysis, indicating their origin as stated above. A 3D visualization showed complex pore structure in the arable vineyard soil, with large vertical and horizontal biopores, confirming the importance of combining various techniques for preferential flow determination. By combining the dye staining and X-ray imaging, different flow patterns were revealed in the soil matrix and macropores. Additional modeling of the transport of Brilliant Blue dye revealed the necessity of using dual-region transport models to capture non-equilibrium flow patterns in agricultural soils with complex pore systems.

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.

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