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Meta-analysis of the effects of soil properties, site factors and experimental conditions on preferential solute transport

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Abstract

Preferential flow is a widespread phenomenon that is known to strongly affect solute transport in soil, but our understanding and knowledge is still poor of the site factors and soil properties that promote it. To investigate these relationships, we assembled a database from the peer-reviewed literature containing information on 793 breakthrough curve experiments under steady-state flow conditions. Most of the collected experiments (642 of the 793 datasets) had been conducted on undisturbed soil columns, although some experiments on repacked soil, clean sands, and glass beads were also included. In addition to the apparent dispersivity, we focused attention on three potential indicators of preferential solute transport, namely the 5 %-arrival time, the holdback factor, and the ratio of piston-flow and average transport velocities. Our results suggest that in contrast to the 5 %-arrival time and the holdback factor, the piston-flow to transport velocity ratio is not related to preferential macropore transport but rather to the exclusion or retardation of the applied tracer. Confirming that the apparent longitudinal dispersivity is positively correlated with the travel distance of the tracer, our results also illustrate that this correlation is refined if the normalized 5 %-tracer arrival time is also taken into account. In particular, we found that the degree of preferential solute transport increases with apparent dispersivity and decreases with travel distance. A similar but weaker relationship was observed between apparent dispersivity, 5 %-tracer arrival time, and lateral observation scale, such that the strength of preferential transport increases with lateral observation scale. However, we also found that the travel distance and the lateral observation scale in the investigated dataset are correlated which makes it difficult to distinguish their influence on these transport characteristics. We observed that anionic tracers exhibited larger apparent dispersivities than electrically neutral tracers under comparable experimental conditions. We also found that the strength of preferential transport increased at larger flow rates and water saturations, which suggests that macropore flow was a more important flow mechanism than heterogeneous flow in the soil matrix. Nevertheless, our data shows that heterogeneous

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flow in the soil matrix also occasionally leads to strong preferential transport. Furthermore, we show that preferential solute transport under steady-state flow depends on soil texture in a threshold-like manner: moderate to strong preferential transport was found to occur only for undisturbed soils which contain more than 8 % clay. Preferential flow characteristics were also absent for columns filled with glass beads, clean sands, or sieved soil. No clear effect of land use on the pattern of solute transport could be discerned, probably because the available dataset was too small and too much affected by cross-correlations with experimental conditions. Our results suggest that in developing pedotransfer functions for solute transport properties of soils it is critically important to account for travel distance, lateral observation scale, and water flow rate and saturation.

1 Introduction

During recent decades the number and quantity of man-made substances that are released onto the soil has been increasing exponentially. Therefore it is becoming more and more important to be able to quantify and predict water and solute fluxes through soil as knowledge of the latter is fundamental to deciding on appropriate prevention or remediation strategies. Quantitatively accurate estimation of water and solute fluxes in soils requires knowledge of hydraulic and solute transport properties. However, their direct measurement is labour-intensive and costly. As they are in most cases also spatially highly variable, it is not possible to measure them directly at a sufficiently high spatial resolution at the relevant scales for management, such as the field, region or landscape scale. Pedotransfer functions (PTFs) offer a way out of this dilemma (Wösten et al., 2001). PTFs denote an approach in which soil properties that are difficult to measure, e.g. the water retention properties, are estimated using other soil properties that are easier to measure, e.g. the bulk density or texture, as proxy variables. Most work so far has focused on soil hydraulic properties, and very little effort has been devoted to developing PTF's for solute transport characteristics. Some approaches for identifying

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“local” PTFs for parameters of the convection-dispersion equation (CDE) or the mobile-immobile model (MIM) have been published based on relatively small datasets (less than 25 samples in all cases) that had been collected explicitly for the purpose (e.g. Goncalves et al., 2001; Perfect et al., 2002; Shaw et al., 2000; Vervoort et al., 1999). In other studies, data from peer-reviewed literature was assembled to construct larger databases of solute breakthrough curve (BTC) experiments (e.g. Rose, 1977; Beven et al., 1993; Griffioen et al., 1998; Oliver and Smettem, 2003). In these studies, the authors investigated correlations among CDE and MIM model parameters of between 50 and 359 BTC experiments, but links to soil properties and experimental conditions were hardly discussed. In contrast, such links were explicitly established in the study by Bromly et al. (2007), who focused on the relationship of a CDE model parameter, the (longitudinal) dispersivity, to properties of saturated repacked soil columns. Their database comprised 291 entries. Another large database of BTC data was published by Vanderborght and Vereecken (2007). It contains 635 datasets of flux and resident concentration BTC experiments with conservative tracers on undisturbed soil and covers all scales between the small column-scale and the field-scale. Vanderborght and Vereecken (2007) used the dataset to investigate how the longitudinal dispersivity is related to scale, boundary conditions, soil texture, and measurement method. They confirmed that the transport distance and the longitudinal dispersivity are generally positively correlated in soils. The same observation had been previously reported for tracer experiments in groundwater (Gelhar et al., 1992; Neuman, 1990).

All of the above discussed studies have “a priori” assumed the validity of one solute transport model, usually the CDE or the MIM. However, it seems likely that no single model is able to properly characterize all of the contrasting flow regimes found in soils, including convective-dispersive transport, heterogeneous flow (funnel flow), non-equilibrium flow in soil macropores or unstable finger flow (Jury and Flühler, 1992). Indeed, it is commonly found that the flow or mixing regime may change one or more times along the travel path (e.g. Vanderborght et al., 2001), as soils are predominantly layered in the horizontal direction and solute transport normally takes place in the

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vertical direction. In effect, a simple generally applicable model for solute transport in soils that is at the same time consistent with the underlying physics is presently not available. Therefore, model-independent (non-parametric) PTFs for solute transport properties should be preferred to model-dependent ones. Some indicator of the strength of preferential transport is then required in place of the model parameters. Several candidates for such an indicator have been proposed during recent years. Among them are the skewness of the BTC (e.g. Stagnitti et al., 2000), the pore volumes drained at the arrival of the peak concentration (Ren et al., 1996; Comegna et al., 1999), the "holdback factor", defined as the amount of original water remaining in a column when one pore volume of displacing water has entered (Danckwerts, 1953; Rose, 1973) and early quantiles of solute arrival times (Knudby and Carrera, 2005).

In this study, we expand and broaden earlier efforts (e.g. Vanderborght and Vereecken, 2007) to develop a database of solute transport experiments derived from the published literature, which comprises a larger number of BTCs ($n = 793$) with accompanying information on soil properties, site factors (e.g. land use and soil management) and experimental conditions. In contrast to Vanderborght and Vereecken (2007) we only included BTC experiments with direct flux concentration measurements to improve comparability of the collected data. Our main motivation for this work was to enable the development of generally applicable non-parametric PTFs for inert solute transport. In this paper, we present the database and the results of initial analyses that relate derived BTC-shape measures as indicators of the degree of preferential transport and dispersion to experimental boundary conditions, soil properties and site factors.

2 Material and methods

We collected information on 793 BTCs for inert tracers in steady-state flow experiments on undisturbed soil samples and from a smaller number of columns filled with glass beads, clean sands, or sieved and repacked soil. The data was taken from 80 articles

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published in the peer-reviewed literature. Details on the data sources are given in Table 1. We deliberately excluded BTCs consisting of resident concentration data (e.g. sampled by time-domain reflectometry) or data from local sampling methods (e.g. suction samplers). Thus, all the considered BTCs were obtained from measurements of flux concentrations in column or tile-drain effluents. Alongside the BTCs, additional information on corresponding soil properties, site factors and experimental conditions was gathered and stored in a relational MySQL database. Table 2 gives an overview on soil properties, site factors and experimental conditions collected in the database as well as information on their completeness.

One difficulty in comparing experimental data is that several different soil texture classification systems were used in the 80 articles. All the classification systems have in common that they assign all particles with an equivalent diameter of less than two micrometers to the clay fraction, but the boundary between the silt and sand fraction varies. We standardized all texture data to the USDA classification system, which sets the silt/sand boundary at 50 µm. We did this by log-linear interpolation (Nemes et al., 1999). For soil columns containing two or more soil layers, we derived an effective soil textural composition by calculating the layer-thickness-weighted average of the sand, silt and clay fractions, respectively. In addition, we computed the geometric mean grain diameter using the approach published in Shirazi et al. (2001).

We used CDE and MIM parameter sets to estimate travel-time PDFs for each BTC. The travel-time PDF of a BTC is equivalent to the breakthrough curve which would have been obtained for a Dirac tracer input. Note that we do not ascribe the PDF to any specific transport process in our study. The only reason for choosing CDE and MIM parameters-sets is the frequent application of these two models in the peer-reviewed literature. By using CDE and MIM parameter-sets, we are able to also include studies in which only MIM or CDE model parameters were reported rather than raw data of the actual BTCs. The 793 BTCs in our database consist of 165 BTCs scanned from raw data, 426 BTCs for which only MIM parameters were available and 202 BTCs for which CDE parameters were published. For the 165 datasets for which the BTC raw

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data was available, MIM parameters were inversely determined by fitting CXTFit 2.1 (Toride et al., 1999, command-line version published as part of the STANMOD package, version 2.07). We included this step to make the 165 datasets with BTC raw data more comparable to the remaining 628 BTCs for which only model parameters were available. A drawback to this approach is that some PDFs are then only reconstructed in an approximate manner due to the limited degrees of freedom of the MIM transfer-function and its inability to fit some of the BTCs. Nevertheless, the MIM and CDE fitted the BTC very well in most cases, with a geometric mean coefficient of determination, R^2, of 0.99. Alternative methods for PDF-reconstruction could be preferable, especially in those few cases where the CDE or MIM did not fit well. For example, the BTCs could be deconvoluted using a mixture of standard-type transfer functions (see e.g. Koestel et al., 2011) or by imposing a smoothness constraint (Skaggs et al., 1998).

We used analytical solutions of the CDE and MIM for Dirac-pulse input, flux concentrations in input and effluent and a semi-infinite domain (Valocchi, 1985) to reconstruct the PDF's. We then derived four non-parametric shape-measures from the reconstructed PDFs (Koestel et al., 2011). Firstly, we investigated the ratio of the piston-flow velocity, v_q (cm d^{-1}), to the average transport velocity, v_n (cm d^{-1}), denoted as η (–) and defined by

$$\eta = \frac{v_q}{v_n} \tag{1}$$

where

$$v_q = \frac{q}{\theta} \tag{2}$$

and

$$v_n = \frac{L}{\mu'_1} \tag{3}$$

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where q (cm d^{-1}) is the water flux, θ is the (total) volumetric water content (–), L is the column length (cm) and μ_1' is the normalized first moment of the PDF,

$$\mu_1' = \frac{m_1}{m_0} \quad (4)$$

where m_0 and m_1 are the zeroth and first moments, respectively, defined as

$$m_0 = \int_0^\infty f\,dt \quad (5)$$

and

$$m_1 = \int_0^\infty t f\,dt \quad (6)$$

where f (t^{-1}) denotes the PDF. The piston-flow to transport velocity ratio, η, is smaller than one if the solute is transported faster than the water and it is larger than one if the solute is retarded relative to the water. It is a non-parametric analogue to the retardation coefficient in the CDE and MIM. Vanderborght and Vereecken (2007) used the reciprocal of η, i.e. $1/\eta$, to investigate preferential transport. They suggested that $\eta < 1$ indicates bypass flow.

The second shape-measure used in this study is the normalized arrival-time of the first five percent of the tracer, $p_{0.05}$ (–). It is derived from the normalized arrival times, T (–),

$$T = \frac{t}{\mu_1'} \quad (7)$$

and the dimensionless PDF, f_n (–),

$$f_n = f\mu_1' \quad (8)$$

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It is easily obtained from the dimensionless cumulative distribution function (CDF), F_n (–), which is calculated by integrating f_n,

$$F_n = \int_0^\infty f_n dT \tag{9}$$

Figure 1 illustrates how $p_{0.05}$ is derived for a BTC taken from Garré et al. (2010). $p_{0.05}$ is bounded by zero and one, where a value of one indicates piston flow and it is negatively correlated with the degree of preferential transport (Knudby and Carrera, 2005).

We also investigated the holdback factor, H (–), as another measure of preferential transport. This was introduced by Danckwerts (1953) to characterize the degree of mixing of two solutes in a vessel:

$$H = \int_0^1 F_n dT \tag{10}$$

It corresponds to the "amount of original fluid remaining in the column when one (water-filled) pore volume of displacing fluid has entered" (Rose, 1973). It follows that a large H indicates preferential characteristics in a transport process. H is calculated as the integral of the dimensionless CDF between zero and one. The holdback factor, H, is also illustrated in Fig. 1. Although it appears to be a convenient indicator of preferential transport, it has so far only been applied occasionally. H has the advantage over $p_{0.05}$ that it samples a larger part of the CDF, but has the disadvantage that it is less robust to the chosen method of BTC-deconvolution (Koestel et al., 2011). We believe that H is superior to $p_{0.05}$ in characterizing preferential behavior from a BTC provided that a long data-series with a high temporal resolution is available.

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Finally, we also investigated the apparent dispersivity, λ_{app} (cm), which is defined as

$$\lambda_{app} = \frac{\mu_2 L}{2} \tag{11}$$

where μ_2 (–) is the second central moment of the dimensionless PDF,

$$\mu_2 = \int_0^\infty (T-1)^2 f_n dT \tag{12}$$

Note that μ_2, as it is defined here, is identical to the coefficient of variation. The apparent dispersivity, λ_{app}, is generally thought to be an indicator of heterogeneity of the solute transport process (Vanderborght and Vereecken, 2007) and thus may complement the above discussed shape-measures. It has the disadvantage that it is less robust to the choice of BTC-deconvolution method than say $p_{0.05}$ (Koestel et al., 2011). One advantage of λ_{app} as a shape measure is that it has already been intensively investigated in the literature (Bromly et al., 2007; Vanderborght and Vereecken, 2007; Hunt and Skinner, 2010).

3 Results and discussion

338 of the 793 experiments available in the database correspond to undisturbed soil samples from arable land (Table 3). 231 of them are from conventionally-tilled fields, 6 from fields with reduced or conservation tillage and 45 from fields with no tillage at all. For the remaining 56 samples, the soil management practices were not specified. Managed or natural grassland is the second most common land use type represented in the database ($n = 121$). Samples with arable and grassland land use are distributed over most of the texture triangle with no apparent bias towards any textural class (see Fig. 2). In contrast, the 80 BTCs from samples from forest sites are restricted to soil samples with less than 25 % clay (Fig. 2). Other land uses, like orchard ($n = 22$),

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grass ley ($n = 7$) or heathland ($n = 2$), are rare. 101 BTCs were measured on samples with unspecified land use. Finally, the 793 datasets also contain 119 experiments on sieved and repacked columns, 32 experiments on columns filled with clean sands or glass beads and 60 experiments on undisturbed samples taken from more than 1 m below the land surface (Table 3). All these studies were conducted on soil columns, except Radcliffe et al. (1996) who sampled the BTCs from tile-drain discharge. Figure 2 illustrates that the majority of the solute transport experiments had been performed on undisturbed but rather short soil columns which had been sampled from one single soil horizon (see also Table 3).

An overview of Spearman rank correlations among the investigated soil properties, experimental conditions, and BTC shape measures is given in Fig. 3. The asterisks indicate p-values of less than 0.001. Some correlations are unsurprising, such as the positive correlations between the flux, q, the average transport velocity, v, the average pressure head, h, and the water content, θ. Other similar examples are the correlations between geometric mean grain diameter, d_g, bulk density, ρ, and clay, silt, and sand fractions. Also, the positive correlation between average sampling depth and the soil sample length (which is identical to the travel distance), L, is easily explained, as sampling pits for larger soil columns must necessarily extend deeper into the ground. Likewise, the column cross-section, A, is positively correlated with L (and the sampling depth).

We found a positive correlation of the apparent dispersivity, λ_{app}, with travel distance, L, and lateral observation scale, A. This confirms what has been in general found in already published reviews on dispersivity (e.g. Gelhar et al., 1992; Vanderborght and Vereecken, 2007), although it is hardly possible to separate the effects of L and A on λ_{app} due to their large mutual correlation. Also consistent with previous studies, Fig. 3 shows a positive correlation between the apparent dispersivity, λ_{app}, and the water flux, q, as well as the pressure head, h. Furthermore, the correlation coefficients with texture data show that λ_{app} was in general larger for finer textured soil and smaller for coarse textures which also is in accordance with empirical knowledge and has also

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been reported by Vanderborght and Vereecken (2007). Finally, we observed no correlation between organic carbon content, OC, and apparent dispersivity, λ_{app}.

Two of the three investigated indicators for preferential transport, namely the normalized 5 %-arrival time, $p_{0.05}$, and the holdback factor, H, were strongly negatively correlated. This confirms the findings of Koestel et al. (2011) on a smaller dataset. According to these two shape-measures, the degree of preferential transport increased with flux, q, pressure head, h, and water content, θ. This is consistent with empirical findings that show that preferential flow and transport are more likely to be observed under saturated and near-saturated conditions (Langner et al, 1999; Seyfried and Rao, 1987). The correlation matrix indicates that the degree of preferential transport was positively correlated with the lateral observation scale, A, but not with the transport distance, L. An intuitive explanation for this is that increasing the lateral observation scale also increases the probability of sampling preferential flow paths, whereas an increase in transport distance decreases the probability of connected preferential flow paths in the transport direction. We consider it likely that a negative correlation between transport distance and preferential transport characteristics was masked by the strong mutual correlation between L and A. Both shape-measures, $p_{0.05}$ and H, indicate a positive correlation between the degree of preferential transport and the clay and silt fraction, and a negative correlation to the geometric mean grain diameter and the sand fraction. Also, a weak negative correlation between the strength of preferential transport and bulk density, ρ, was found, but no correlation to the organic carbon content, OC.

The fourth shape-measure, the piston-flow to transport velocity ratio, η, was not significantly correlated to either the normalized 5 %-arrival time, $p_{0.05}$, or to the apparent dispersivity, λ_{app}. A very weak positive correlation was found between η and the holdback factor H. Moreover, we observed that solute transport was increasingly retarded ($\eta > 1$) with increasing water flow rate, q, and pressure heads, h. We found no significant correlations between η and any of the investigated soil properties (i.e. geometric mean grain diameter, d_g, bulk density, ρ, texture fractions and organic carbon content, OC). It follows that the piston-flow to transport velocity ratio, η, reflects different information on solute transport characteristics as compared to the preferential transport indicators, $p_{0.05}$ and H.

Figure 4a shows that strong correlation between the 5 %-arrival time, $p_{0.05}$, and the holdback factor, H, was weaker for small $p_{0.05}$ (large H), i.e. for BTCs displaying strong preferential transport. Figure 4a suggests that H offers a better discrimination between soils showing strong preferential transport whereas $p_{0.05}$ better resolves differences among soils with weaker preferential transport characteristics. In Fig. 4b and c, the piston-flow to transport velocity ratio, η, is compared to $p_{0.05}$ and H. Note that no value for η was available if no independent water content measurement was published for the respective BTC (see Eq. 2). Therefore, the range of $p_{0.05}$ in Fig. 4b appears to be different to the one in Fig. 4a. Besides depicting the minimal correlation of η to the other two indicators for preferential transport, these two figures also illustrate that η was, in contrast to $p_{0.05}$ and H, sensitive to the choice of tracer in the BTC experiments. Anionic tracers like chloride and bromide were generally transported faster than the water flux whereas the electrically neutral tracers deuterium and tritium only occasionally exhibited accelerated transport, namely when small $p_{0.05}$ and medium H indicated preferential flow. As we only considered experiments where the anionic tracers were applied on soils with electrically neutral or predominantly negatively charged media, the generally accelerated solute transport for anionic tracers is well explained by anion exclusion (Rose et al., 2009; Thomas and Swoboda, 1970). Notably, for very strong preferential transport ($p_{0.05} < 0.1$ and $H > 0.4$), the anionic tracers were retarded.

Figure 5 illustrates the impact of the choice of tracer on BTCs. The non-ionic tracers tritium and deuterium were generally used on longer columns than chloride and bromide and under similar water fluxes. Although longer columns should lead to larger apparent dispersivities, λ_{app} (Fig. 3), this was not observed for the BTCs obtained with tritium and deuterium. This supports the validity of model approaches in which the solute dispersivity is not only dependent on the pore-space geometry but also on the adsorptive properties of tracer and soil matrix (Wels et al., 1997; Pot and Genty, 2007).

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In addition, the strength of preferential transport, as expressed by $p_{0.05}$, was smaller for the non-ionic tracers than for the anions. However, in this case it is more difficult to separate the influences of electrical charge, water flux and column length.

Figure 6a illustrates that for a given value of λ_{app}, $p_{0.05}$ increases with the column length, L. This suggests that the strength of preferential transport decreases with travel distance. No significant linear correlation was found between L and $p_{0.05}$ (Fig. 3), probably because it was masked by the non-linearity of the ternary relationship between L, $p_{0.05}$ and λ_{app}, especially for strong preferential transport ($p_{0.05} < 0.1$). Thus, including $p_{0.05}$ into a scaling-scheme for the apparent dispersivity, λ_{app}, with travel distance, L, strongly increases the amount of explained variance. The first two principal components for the three measures $\log_{10} L$, $\log_{10} \lambda_{app}$ and $p_{0.05}$ (normalized to a mean of zero and a standard deviation of one) explain 91.9 % of the variance between the three shape-measures. In contrast, the first principal component of just $\log_{10} \lambda_{app}$ and $\log_{10} L$ explains only 65.2 % of the variance, exhibiting a Spearman rank correlation coefficient of 0.35 (p-value < 0.001). A very similar ternary relationship was found between $\log_{10} \lambda_{app}$, $p_{0.05}$, and the logarithm of the area of the breakthrough plane, $\log_{10} A$ (Fig. 6b), which explained 88.7 % of the inherent variance. The first principal component between only λ_{app} and A explains 65.1 % of the variance. The corresponding Spearman rank correlation coefficient is 0.47 (p-value < 0.001).

Figure 7a–d shows the dependency of v, λ_{app}, $p_{0.05}$, and η on water flow rates. Only undisturbed samples were considered. Figure 7a–c shows that not only the medians of v and λ_{app} monotonously increase with the respective water flux class but also the strength of preferential transport (there is negative relationship between $p_{0.05}$ and q). This suggests that, for this dataset, macropore transport overshadows preferential transport caused by heterogeneities in matrix hydraulic properties. Nevertheless, Fig. 7c also illustrates that preferential transport cannot be completely ruled out for small water fluxes. Little dependence of the piston-flow to transport velocity ratios, η, on the water flux, q, is observed (Fig. 7d). This indicates that η is not related to preferential transport in soil macropores. Indeed, η is smallest for the experiments with the lowest water fluxes. As most of the experiments included in this analysis were conducted with anionic tracers, a possible explanation for this behavior is that anion exclusion was amplified for experiments under small water flow rates which by trend correspond to experiments under far from saturated conditions when only meso- and micropores are water-filled.

Figure 8 depicts how the soil horizon from which the sample had been taken is related to λ_{app} and $p_{0.05}$. Firstly, Fig. 8 illustrates that samples that contain both topsoil and subsoil exhibit larger apparent dispersivities, λ_{app}, than samples from only topsoil or only subsoil. One obvious explanation for this is that samples containing both topsoil and subsoil are generally longer, so that λ_{app} is also larger due to its positive correlation with travel distance (see Fig. 3). However, it is also plausible that features at the interfaces between topsoil and subsoil in these columns, e.g. plow pans, enhance the spreading of a solute plume, such as observed for example by Öhrström et al. (2002) and Koestel et al. (2009b). As samples taken from only the topsoil are always restricted to lengths between 20 and 40 cm and because longer samples taken from only the subsoil have seldom been investigated, it is not possible to appraise to what degree interfaces between topsoil and subsoil add to the scaling effect of the apparent dispersivity, λ_{app}, with travel distance. Furthermore, soil columns filled with clean sands or glass beads, which are tagged as "irrelevant" in Fig. 8, generated strictly non-preferential BTCs.

The relationship between λ_{app} and $p_{0.05}$ and soil texture, characterized by the geometric mean grain diameter, d_g, is somewhat more complicated (see Fig. 9). Coarser-textured soils with large d_g are not at all restricted to a specific range of apparent dispersivities or 5 %-arrival times, or specific combinations of the two. In contrast, for fine-grained soils, $p_{0.05}$ is always less than 0.6 and the apparent dispersivity always exceeds ca. 2 cm. Finally, the samples with an intermediate d_g show low λ_{app}-to-$p_{0.05}$ratios upon visual inspection (Fig. 9). Such a ratio is also typical for short transport distances (Fig. 3). A possible explanation may be that in our dataset, experiments on soils with intermediate d_g were only carried out on short columns. In summary, there

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are no smooth transitions apparent in Fig. 9 and the geometric mean grain diameter appears not to be a strong predictor for λ_{app} and $p_{0.05}$.

A clearer picture emerges if λ_{app} and $p_{0.05}$ are plotted in relation to USDA texture classes. Figure 10a shows that BTCs showing strong preferential transport characteristics ($p_{0.05} < 0.2$) are restricted to samples containing at least 8 to 9 % clay. This is similar to the clay content needed for the formation of stable soil aggregates (Horn et al., 1994) and may also reflect an absence of biopores in such soils, since both roots and earthworms avoid coarse single-grain soils. Also, small $p_{0.05}$ values are less common for samples with more than 50 % silt. However, the latter may possibly be an artifact caused by the scarcity of experiments on short columns sampled from just one single soil horizon in silty soils (see Fig. 10a–d). The apparent dispersivity, λ_{app}, roughly follows the distribution of $p_{0.05}$ on the texture triangle diagram (Fig. 10b) which is not surprising given the strong correlation between the two (see Fig. 3). However, extreme λ_{app} values were less clearly constrained to specific regions on the texture triangle diagram. They mostly occurred for undisturbed samples containing more than one soil horizon. Finally, Fig. 10c shows the distribution of the piston-flow to transport velocity ratio, η on the texture triangle. Small piston-flow to transport velocity ratios ($\eta \ll 1$), were predominantly found for loamy soils and were absent for soils in which one of the three fractions (silt, sand or clay) dominates. The complete absence of $\eta < 1$ for soils of clayey texture may be related to anion exclusion as all these experiments were conducted with anionic tracers (see Table 1 and discussion above). Small η occur exclusively in loamy soils which are characterized by a broader particle (and thus pore) size distribution than soils from other texture classes. As a broader pore size spectrum should enhance heterogeneous transport in the soil matrix, it is possible that, in addition to anion exclusion, η reflects heterogeneous transport in the matrix rather than macropore flow.

Finally, we also investigated the relationships of the BTC shape-measures λ_{app} and $p_{0.05}$ with land use and soil management practices. Figure 11a and b illustrates that the 642 undisturbed soil samples exhibited a median apparent dispersivity of 7.3 cm and a median normalized 5 %-arrival time of 0.28 corresponding to steady state flow conditions with a median flux of 12.7 cm d^{-1} and a median travel distance of 23 cm. Much smaller $p_{0.05}$ values were only found for samples from arable sites with reduced tillage and grass leys (Fig. 11a). However, the number of samples for these land use classes was very small, while Fig. 11b reveals that the experiments were conducted on relatively short columns and large water fluxes, both of which promote low $p_{0.05}$. Similarly, the experimental conditions were also not representative for the bulk of the experiments on undisturbed samples for the "forest" sites. For these samples, the experimental conditions promoted larger $p_{0.05}$ values (Fig. 11b). Figure 11a and b shows that sieved and repacked soil samples resulted in clearly larger $p_{0.05}$ values than samples of undisturbed soil, even though the experimental conditions favored small values. The lack of preferential transport for the disturbed samples is consistent with the destruction of natural well-connected pore-structures by sieving. This furthermore underlines the importance of conducting leaching studies on undisturbed samples (see also Elrick and French, 1966; Cassel et al., 1974; McMahon and Thomas, 1974). Furthermore, no sign of preferential transport was found for the BTCs collected from artificial porous media like clean sand or glass beads. They exhibited extremely large $p_{0.05}$ and extremely small λ_{app}, although the experimental conditions should have acted in the opposite direction. Of the natural soils, only the two samples from heathland sites consisting almost of pure sand (Seuntjens et al., 2001) show similar features (Fig. 11a). We conclude that, with a few exceptions, a complete absence of preferential characteristics in solute transport is only observed in artificial homogeneous porous media. Apart from this, our data does not show any clear relationship between land use and degree of preferential transport and solute dispersion. However, such relationships cannot be ruled out, since in our dataset they may have been obscured by a lack of comparable experimental conditions.

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4 Conclusions

We investigated the controls on inert solute transport based on 793 breakthrough curve experiments collected from the peer-reviewed literature, mostly conducted on undisturbed soil columns. We focused especially on four breakthrough curve shape-measures, namely the normalized 5 %-arrival time, the holdback factor, the apparent longitudinal dispersivity and the ratio of piston-flow and average transport velocities. The normalized 5 %-arrival time, the dispersivity and the holdback factor were strongly correlated, while only weak correlations were found between these shape-measures and the piston-flow to transport velocity ratio, suggesting that the latter contains complementary information on solute transport. In particular, our results suggest that the piston-flow to transport velocity ratio is more strongly related to exclusion or retardation of the applied tracer and preferential transport in the soil matrix, rather than to the degree of preferential solute transport in macropores.

Our results indicate that not only the transport velocity but also the apparent dispersivity is dependent on the choice of tracer. Anionic tracers exhibited larger apparent dispersivities than electrically neutral ones. Moreover, our results confirm the findings of previous studies that the apparent longitudinal dispersivity is positively correlated with the travel distance of the tracer. We found that this relationship is refined if the normalized 5 % tracer arrival time is also taken into account as a measure of the degree of preferential solute transport. In particular, we found that the degree of preferential solute transport increases with apparent dispersivity and decreases with travel distance. A similar relationship was found between the apparent dispersivity and the lateral observation scale. However, the effects of travel distance and lateral observation scale on these two measures are difficult to separate as travel distance and breakthrough plane cross-sectional area were positively correlated.

The strength of preferential transport increased at larger flow rates and water saturations, which suggests that macropore flow was a dominant flow mechanism for the experiments in our database. Nevertheless, our data shows that heterogeneous flow

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in the soil matrix also occasionally leads to strong preferential transport characteristics, especially in loamy soils. It should also be noted here that most of the studies included in the database were conducted under relatively high intensity and steady-state irrigation boundary conditions and saturated or near-saturated initial conditions. Therefore, the general relevance of transport processes that are triggered under different initial and/or boundary conditions cannot be investigated with our database. Examples are unstable finger flow (Scheidegger, 1960; Raats, 1973; Hendrickx et al., 1993) and preferential transport due to soil hydrophobicity (Thomas et al., 1973; Ritsema and Dekker, 1996) or air-entrapment (Debacker, 1967; Snehota et al., 2008). These flow and transport phenomena have been frequently investigated, but mostly with aid of dye tracers and only occasionally by means of BTC experiments. The lack of appropriate studies to quantify the importance of these preferential transport processes as compared to the here investigated BTC experiments should be addressed in the future.

Preferential solute transport was shown to depend on soil texture in a threshold-like manner: moderate to strong preferential transport was only found in soils with a texture consisting of more than 8 to 9 % clay. As expected, columns filled with glass beads, clean sands, or sieved soil exhibited no preferential transport. No clear effect of land use on the pattern of solute transport could be discerned. However, we suspect that the dataset was too small and also too strongly influenced by cross-correlations with soil type and experimental conditions to allow any firm conclusions to be drawn on this.

The database opens up the possibility to develop pedotransfer functions for solute transport properties in soil. Whilst they are generally encouraging, the results of the initial analyses presented in this paper suggest that this will be a challenging task. In particular, it will be critically important to distinguish the effects of experimental conditions (column dimensions, initial and boundary conditions) from the effects of soil and site characteristics. Some initial attempts in this direction are underway.

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Table 1. Primary source publication and other information on the BTC experiments collected in the meta-database.

Primary reference	# of BTCs	Tracer	Type of soil or porous medium	USDA texture class	Undist. sample?	Land use
Akhtar et al. (2003)	9	chloride	lamellic hapludalf**, glossaquic hapludalf**, fluventic eutrudept**, glossic hapludalf**, typic fragiudept**	loamy sand, loam, silt loam	yes	unknown
Anamosa et al. (1990)	6	tritium	typic gibbsiorthox**	unknown	yes	arable
Barizon (2004)	3	chloride	unknown	sandy loam, sandy clay loam, clay	no	unknown
Bedmar et al. (2008)	6	bromide	unknown	silt loam, silty clay loam	yes	arable
Bromly and Hinz (2004)	14	lissamine FF	clean sand	sand	no	irrelevant
Candela et al. (2007)	7	bromide	typic xerorthent**	silt loam	no	unknown
Coats and Smith (1964)	2	calcium	alundum		no	irrelevant
Comegna et al. (1999)	3	chloride	entisol*, vertisol*, andosol*	sand, clay loam, sandy loam	yes	arable
Comegna et al. (2001)	17	chloride	orchard, arable	silt loam, silty clay loam	yes	unknown
de Smedt and Wierenga (1984)	13	chloride	glassbeads	sand	no	irrelevant
Dousset et al. (2004)	6	bromide	gleyic luvisol*	silty clay loam	yes, no	grass ley
Dufey et al. (1982)	10	chloride	unknown	sandy loam	no	unknown
Dyson and White (1987)	1	chloride	calcaric cambisol*	sandy clay loam	yes	managed grassland
Dyson and White (1989)	17	chloride	calcaric cambisol*	sandy clay loam	yes	managed grassland
Elrick and French (1966)	2	chloride	unknown	loam, silt loam	yes, no	unknown
Ersahin et al. (2002)	12	bromide	mollic planosol*	silt loam	yes	natural grassland
Gaber et al. (1995)	4	tritium	typic haploboroll**	silty clay loam	yes	unknown
Garré et al. (2010)	2	chloride	orthic luvisol*	silt loam	yes	arable
Gaston et al. (2007)	4	bromide	thermic ochraqualf**	clay loam	yes	arable
Gaston and Locke (1996)	4	bromide	thermic ochraqualf**	clay loam	yes	arable

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Table 1. Continued.

Primary reference	# of BTCs	Tracer	Type of soil or porous medium	USDA texture class	Undist. sample?	Land use
Gaston and Locke (2000)	4	bromide	thermic ochraqualf**	clay loam	yes	arable
Goncalves et al. (2001)	24	chloride	dystric fluvisol*, calcic vertisol*, calcaric cambisol*, vertic luvisol*	loam, clay, clay loam, sandy clay loam, sandy loam, sandy clay	yes	arable, orchard
Green et al. (1995)	12	chloride	Clarion silt loam		yes	arable
Gwo et al. (1995)	3	bromide	unknown	unknown	yes	forest
Haws et al. (2004)	5	bromide	mesic typic endoquoll**	silt loam	yes	arable
Helmke et al. (2005)	24	bromide PFBA, PIPES	typic hapludoll**, typic hapludalf**	loam, clay loam, sandy loam	yes	irrelevant
Jacobsen et al. (1992)	10	tritium, chloride	orthic haplohumod**	loamy sand	yes	unknown
Javaux and Vanclooster (2003)	9	chloride	unconsolidated bedrock	sand	yes	irrelevant
Jensen et al. (1996)	19	chloride	unknown	sandy loam	yes	arable
Jensen et al. (1998)	2	tritium	aeric glossaqualf**	sandy loam	yes	arable
Jorgensen et al. (2004)	4	bromide	unknown	sandy loam, sandy clay loam	yes	arable
Kamra et al. (2001)	47	bromide	unknown	sandy loam,	yes	arable, forest
Kasteel et al. (2000)	1	bromide	orthic luvisol*	silt loam	yes	arable
Kim et al. (2007)	7	bromide	unknown	unknown	no	unknown
Kjaergaard et al. (2004)	34	tritium	stagnic luvisol*	sandy loam, sandy clay loam, clay	yes	arable
Koestel et al. (2009a)	4	chloride	gleyic cambisol*	loamy sand	yes	Arable
Krupp and Elrick (1968)	5	chloride	glassbeads	sand	no	irrelevant
Langner et al. (1999)	19	PFBA	typic haploboroll**	unknown	yes	managed grassland
Lee et al. (2000)	3	chloride	stagnosol*	unknown	yes	arable
Lee et al. (2001)	4	bromide	stagnosol*	loam	yes	arable

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Table 1. Continued.

Primary reference	# of BTCs	Tracer	Type of soil or porous medium	USDA texture class	Undist. sample?	Land use
Lennartz et al. (2008)	4	bromide	rendzik leptosol*	silt loam	yes	arable
Luo et al. (2010)	8	bromide	mesic typic hapludalf**	silt loam	yes	arable, managed grassland
Maraqa et al. (1997)	33	tritium	typic udipsamment**, entic haplaquod**	sand	no	unknown
Mayes et al. (2003)	29	bromide PFBA, PIPES	acidic Inceptisol**, unconsolidated bedrock	silt loam, sandy loam	yes	unknown, forest
McIntosh et al. (1999)	4	bromide, chloride	thermic typic dystrochrept**, thermic kanhapludult**	sandy clay loam sandy loam	yes	forest
Montoya et al. (2006)	24	bromide	typic argiudoll**	clay loam, loam	yes	arable
Mooney and Morris (2008)	4	chloride	gleyic luvisol*, cambisol*, gleysol*	sandy loam, clay loam, clay	yes	arable
Nkedi-Kizza et al. (1983)	34	tritium, chloride	oxisol* (sieved aggregates)	sandy loam	no	irrelevant
Oliver and Smettem (2003)	13	bromide	typic xeric psamment**	unknown	no	unknown
Pang et al. (2008)	29	bromide	typic endoaquept**, typic haplohumult**, typic dystrudept**, aeric fragiaquept**, fluventic eutrudept**, typic udipsamment**, typic udivitrand**, typic hapludand**	clay, silty clay, silt loam, sand, sandy loam, loam	yes	managed grassland
Perfect et al. (2002)	2	chloride	typic udifluvent**, vertic endoaquept**	unknown	yes	managed grassland
Pot et al. (2005)	5	bromide	stagnosol*	silt loam	yes	managed grassland
Poulsen et al. (2006)	33	tritium	typic hapludalf**	sandy loam	yes	arable
Prado et al. (2006)	3	deuterium	pachic andosol*	silt loam	no	arable
Prado et al. (2009)	9	deuterium	pachic andosol*	silt loam	yes	arable
Radcliffe et al. (1996)	10	chloride	thermic typic kanhapludult**	clay, clay loam	yes	arable
Raturi et al. (2001)	7	bromide	antroposol*	loamy sand	yes	managed grassland

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Table 1. Continued.

Primary reference	# of BTCs	Tracer	Type of soil or porous medium	USDA texture class	Undist. sample?	Land use
Ren et al. (1996)	20	bromide	durixerollic calciorthid**	silt loam	yes	arable
Reungsang et al. (2001)	12	bromide	typic haploaquoll**, cumulic haploaquoll**	sandy loam	yes	managed grassland, arable
Scherr (2009)	3	bromide	unknown	silty clay loam	yes	managed grassland
Schoen et al. (1999)	3	bromide, chloride, deuterium	unknown	silt loam	yes	arable
Schulin et al. (1987)	23	tritium, bromide	rendzik leptosol*	loam	yes	forest
Segal et al. (2009)	1	bromide	unknown	unknown	yes	arable
Selim and Amacher (1988)	3	tritium	arguic fragiudalf**, typic hapludalf**, typic udipsamment**	unknown	no	unknown
Seo and Lee (2005)	3	chloride	typic hapludult**	sandy loam	yes	unknown
Seuntjens et al. (2001)	2	chloride	podsol*	sand	yes	heathland
Seyfried and Rao (1987)	14	tritium	typic distropept**	unknown	yes	arable, orchard
Shaw et al. (2000)	13	bromide	kandiudult**	sand, sandy loam, loamy sand, sandy clay loam	yes	arable
Singh and Kanwar (1991)	6	chloride	mesic hapludoll**	unknown	yes	arable
Smettem et al. (1983)	3	tritium	unknown	clay loam	yes	arable
Smettem (1984)	12	tritium	"well structured brown calcareous earth"	silt loam	yes	forest
Stagnitti et al. (2000)	1	chloride	unknown	unknown	yes	managed grassland
Tyler and Thomas (1981)	3	chloride	fluventic haplodoll**, typic udifluvent**, vertic haplaquept**	silt loam, silty clay loam, sandy loam	yes	arable
Unold et al. (2009)	4	chloride	orthic luvisol*, gleyic cambisol*	silt loam, sandy loam	yes	arable

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Table 1. Continued.

Primary reference	# of BTCs	Tracer	Type of soil or porous medium	USDA texture class	Undist. sample?	Land use
Vanderborght et al. (2002)	2	chloride	stagnic cambisol*	clay loam	yes	forest
Vervoort et al. (1999)	7	bromide, chloride	typic kandiudult**	sandy loam, sandy clay loam, clay,	yes	managed grassland
Vincent et al. (2007)	8	bromide	stagnosol*	sandy clay loam, silt loam	yes	arable, managed grassland, forest
Vogeler et al. (2006)	12	bromide, chloride	stagnic luvisol*	sandy loam	yes	arable
Wilson et al. (1998)	3	bromide	typic paleudalf**	silt loam	yes	arable
Zurmühl (1998)	2	tritium	unknown	sand	yes	forest

* Classification according to the World Reference Base (WRB).
** Classification according to the system of the United States Department of Agriculture (USDA).

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Table 2. Inventory of the data available in the database.

Data	Available	Missing
Explicit information on water content, q (cm^3 cm^{-3})	504	289
Explicit information on water flux, q (cm d^{-1})	596	197
Travel distance, L (cm)	793	0
Area of breakthrough plane, A (cm^2)	793	0
Information on tracer detection method	793	0
Information on initial conditions	791	2
Pressure head at upper boundary, h_{UB} (cm)	367	426
Pressure head at lower boundary, h_{LB} (cm)	472	321
Average pressure head, h_{ave} (cm)	506	287
Hydraulic gradient, dH/L (–)	433	360
Information on Irrigation device	765	28
Information on outlet construction	752	41
Information on tracer	793	0
Information on tracer application method	793	0
BTC raw data	165	628
Information on land use	675	118
Information on cropping	475	318
Information on soil management practices	400	393
Depth from which soil sample was collected (cm)	563	230
Texture data	670	123
Bulk density, ρ (g cm^{-3})	661	132
Organic carbon content, OC (–)	534	259
Porosity, ϕ (cm^3 cm^{-3})	667	126

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Table 3. Land use and soil management for the 793 datasets in the database.

Land use	# of entries in the database
arable (all)	338
arable (conventional tillage)	231
arable (reduced tillage)	6
arable (no tillage)	45
arable (no further information)	56
forest	80
managed grassland	109
natural grassland	12
grass ley	7
heathland	2
orchard	22
unknown land use	101
sieved and repacked samples*	119
unconsolidated bedrock	39
clean sand or glass beads	32

* Note that for some of the sieved samples the land use was known.

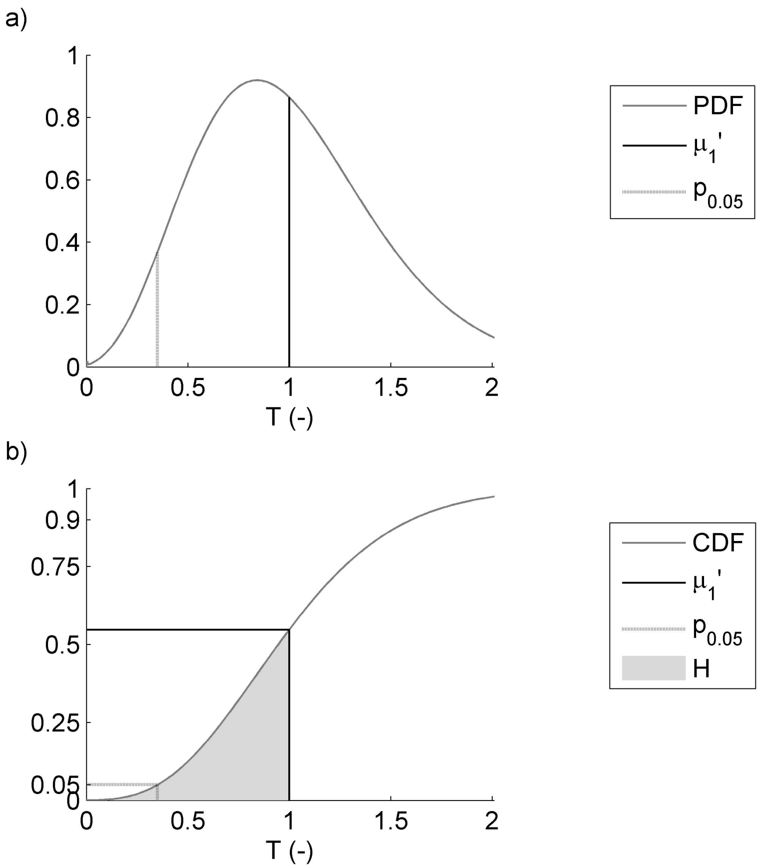

Fig. 1. Normalized PDF **(a)** and CDF **(b)** of an example BTC taken from Garré et al. (2010) illustrating how the normalized first temporal moment, μ_1', the normalized 5 %-arrival time, $p_{0.05}$, and the holdback, H, are derived.

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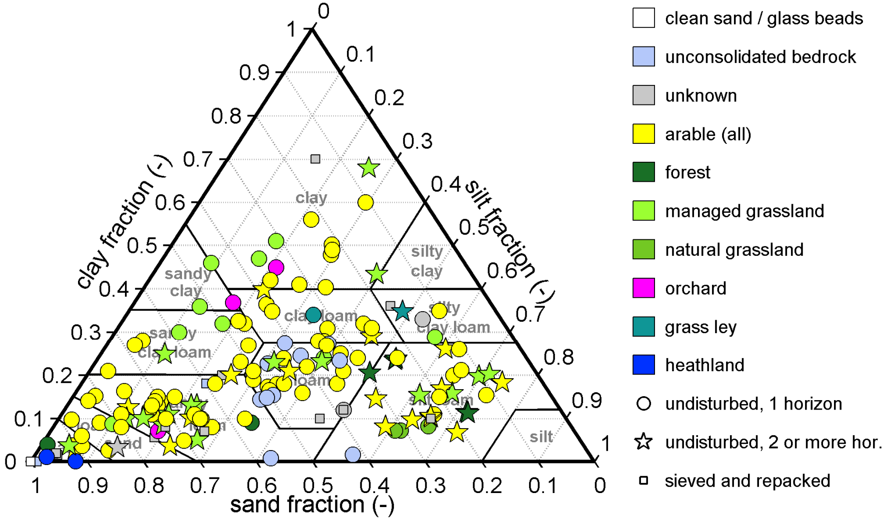

Fig. 2. Land uses corresponding to the soil samples on which the 793 considered BTC experiment had been carried out. Note that in most publications only average values are published for several soil samples and that several experiments are often conducted on one and the same soil sample under different hydraulic boundary conditions. Therefore, the number of datasets visible in the texture triangle is less than 793.

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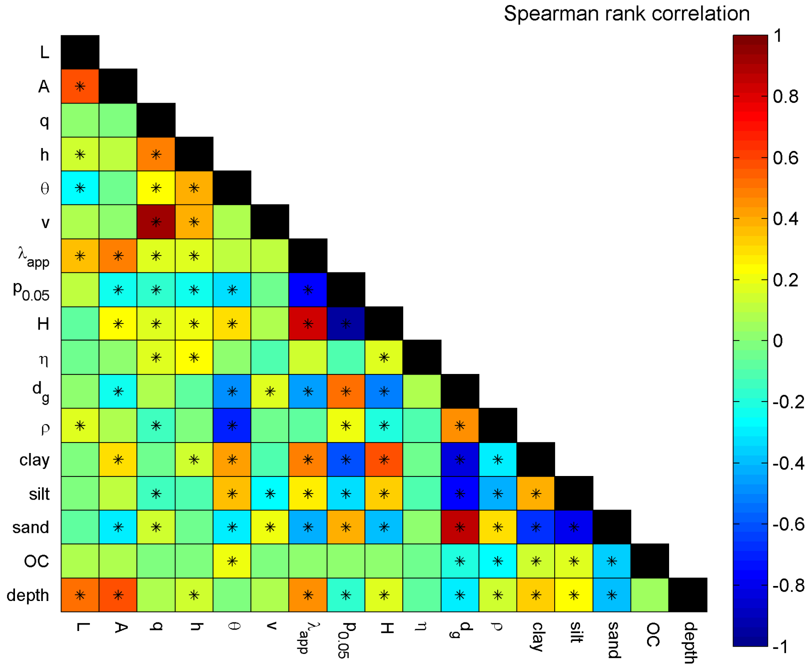

Fig. 3. Spearman rank correlation coefficients between various BTC-shape measures and soil and site as well as experimental properties. The boxes marked by an asterisk indicate significant correlations with p-values of smaller than 0.001. The correlations were carried out for the travel distance, L, the area of the breakthrough plane, A, the water flux, q, the suction head, h, the water content, θ, the transport velocity, v, the apparent dispersivity, λ_{app}, the normalized 5 %-arrival time, $p_{0.05}$, the holdback, H, the piston-flow to transport velocity ratio, η, the geometric mean grain diameter, d_g, the soil bulk density, ρ, the clay fraction, clay, the silt fraction, silt, the sand fraction, sand, the organic carbon content, OC, and the average sampling depth.

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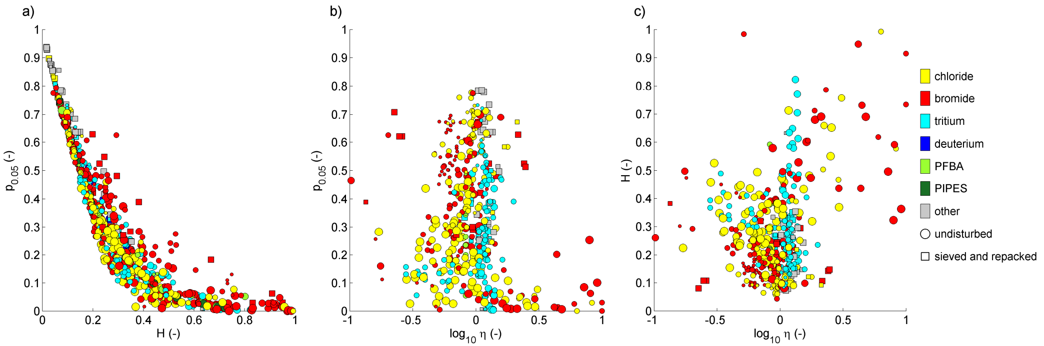

Fig. 4. Comparison between the shape-measures related to the preferential character of the BTC: **(a)** comparison between the holdback, H, and the normalized 5 %-arrival time, $p_{0.05}$; **(b)** comparison of the piston-flow to transport velocity ratio, η, and the normalized 5 %-arrival time, $p_{0.05}$; **(c)** comparison of the piston-flow to transport velocity ratio, η, and the holdback, H. In addition, the type of applied tracer is depicted. The symbol size corresponds to the water fluxes, q, under which the experiment was conducted, small symbols indicating small water fluxes, large symbols denoting large water fluxes.

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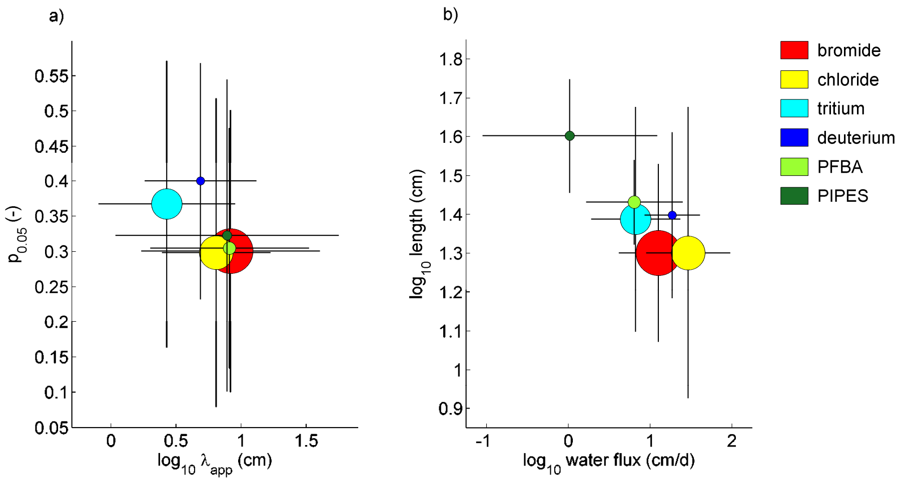

Fig. 5. The median apparent dispersivity, λ_{app}, and normalized 5 %-arrival time, $p_{0.05}$, in dependence of the applied tracer **(a)** and the corresponding median experimental conditions **(b)**. The center of each circle depicts the respective median value and the error bounds indicate the corresponding interquartile range. The size of each circle corresponds to the number of BTC conducted with the respective tracer.

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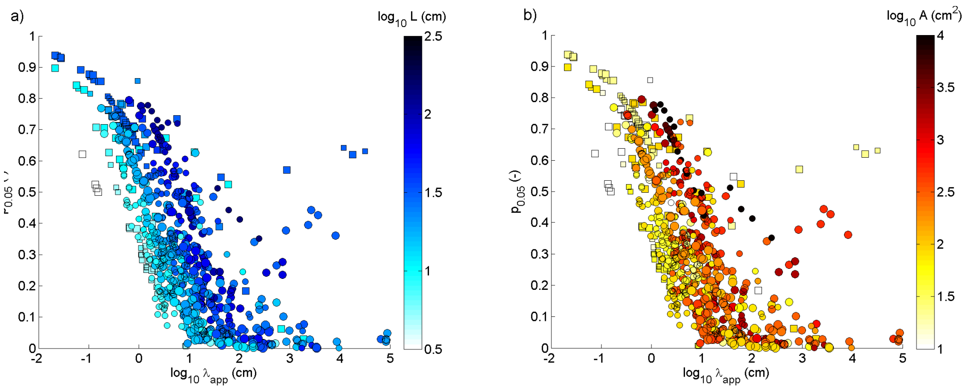

Fig. 6. Comparison of the apparent dispersivity, λ_{app}, and normalized 5 %-arrival time, $p_{0.05}$, with **(a)** the travel distance, L, and **(b)** the area of the breakthrough plane, A. The symbol size corresponds to the water fluxes, q, under which the respective experiment was conducted, small symbols indicating small water fluxes, large symbols denoting large water fluxes. The meaning of the symbol shape is explained in Fig. 4.

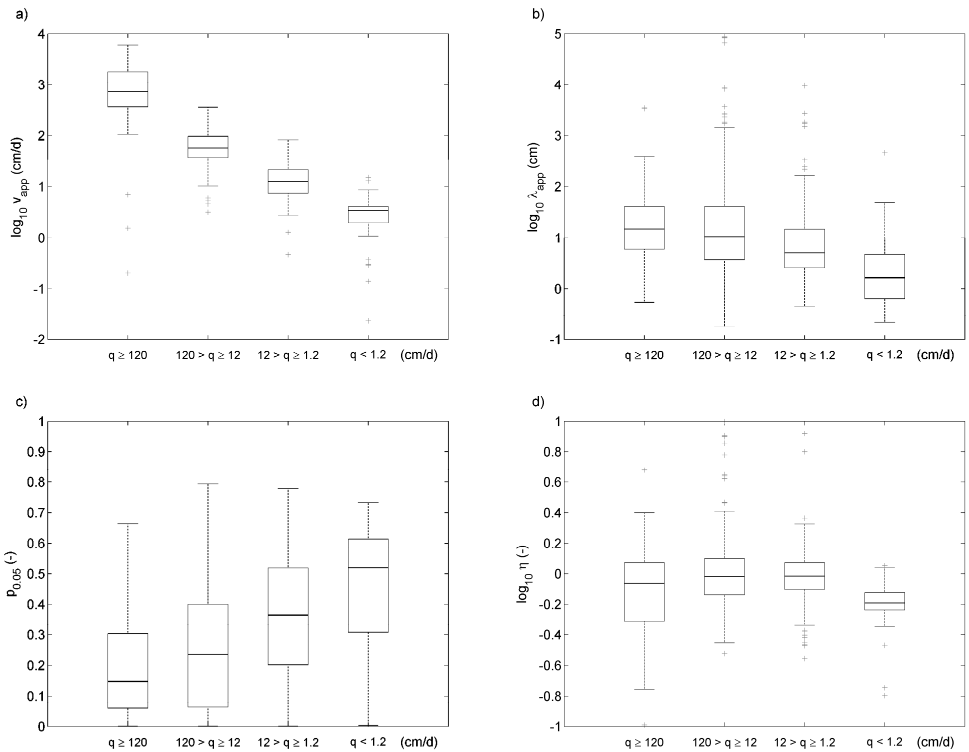

Fig. 7. Boxplots **(a)** transport velocity, v, **(b)** apparent dispersivity, λ_{app}, **(c)** normalized 5 %-arrival time, $p_{0.05}$, and **(d)** piston-flow to transport velocity ratio, η according to the respective water flux class. Note that this figure is based on BTCs from undisturbed soil samples, only.

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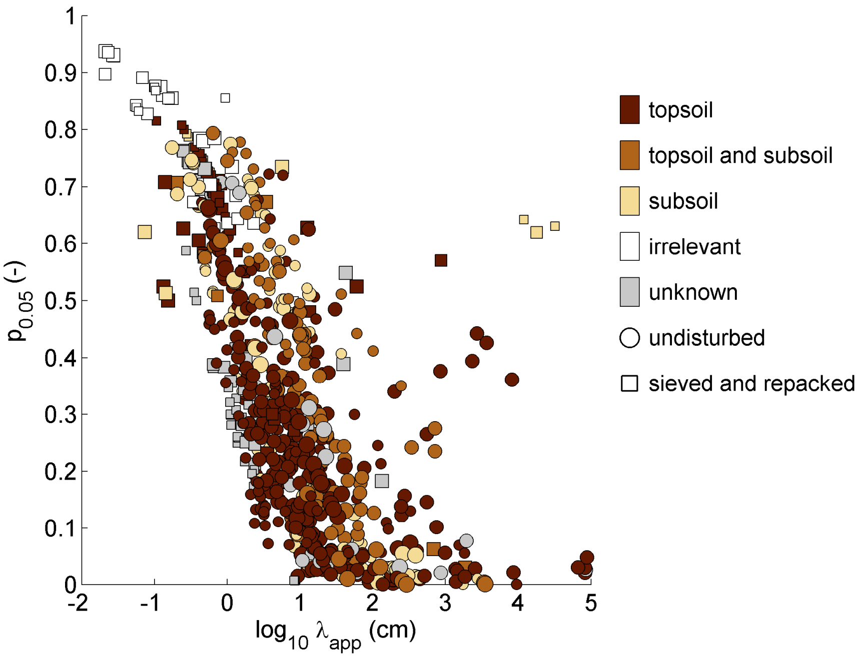

Fig. 8. Comparison of the apparent dispersivity, λ_{app}, and normalized 5 %-arrival time, $p_{0.05}$, with sampling location of the respective soil sample. The symbol size corresponds to the water fluxes, q, under which the respective experiment was conducted, small symbols indicating small water fluxes, large symbols denoting large water fluxes.

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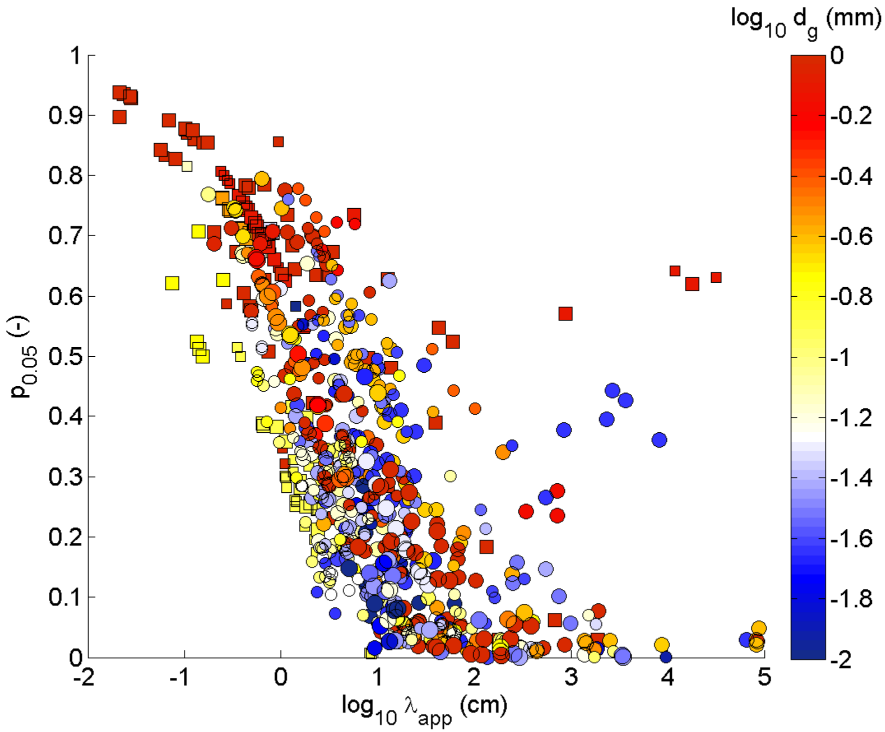

Fig. 9. Comparison of the apparent dispersivity, λ_{app}, and normalized 5 %-arrival time, $p_{0.05}$, with the geometric mean grain diameter, d_g, of the respective soil sample. The symbol size corresponds to the water fluxes, q, under which the respective experiment was conducted, small symbols indicating small water fluxes, large symbols denoting large water fluxes. The meaning of the symbol shape is explained in Fig. 4.

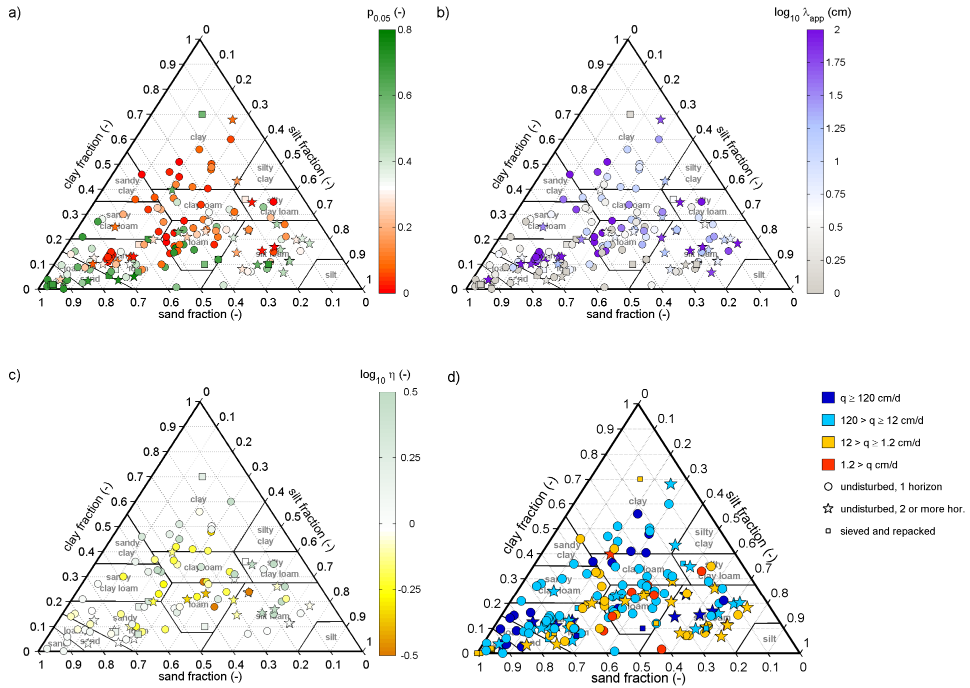

Fig. 10. The **(a)** normalized 5 %-arrival time, $p_{0.05}$; **(b)** apparent dispersivity, λ_{app}; **(c)** the piston-flow to transport velocity ratio, η; and **(d)** the water flux classes corresponding to the considered BTC experiments.

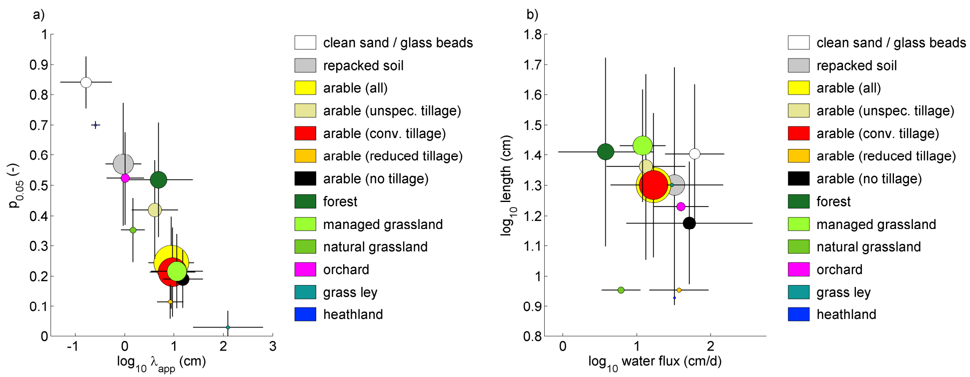

Fig. 11. **(a)** Comparison of the apparent dispersivity, λ_{app}, and normalized 5 %-arrival time, $p_{0.05}$, with the respective land use; **(b)** comparison of the water flux, q, and column length, L, with the respective land use. The center of each circle depicts the respective median value and the error bounds indicate the corresponding interquartile range. The size of each circle corresponds to the number of samples within each land use class.

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