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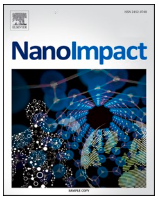

# A meta-analysis of nanomaterial and nanoplastic fate in small column experiments and implications for fate in soils[☆,☆☆]

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ABSTRACT

A long list of possible processes may simultaneously control retention of engineered nanomaterials (NMs) and nanoplastics (NPs) in soils, but there is little insight in which of these processes dominates and under which circumstances. Though not easily transferable to field situations, repacked saturated column tests compose the richest available dataset to explore for overall trends in the behaviour of NMs and NPs in soils. Global attachment efficiencies ($\alpha_{\text{global}}$) were calculated uniformly from metadata of 624 column experiments and correlated against metadata using partial least squares and linear regression analysis. $\alpha_{\text{global}}$ values appeared to some extent operationally defined as they correlate with the experimental column flow rate and in some cases with the particle concentration used in the feedstock. Particle aggregation occurred more as the feedstock concentration increased, but this only had a limited effect on subsequent column retention. In homogeneous sandy media, attachment of particles obeyed well-known trends indicative of non-favourable electrostatic interactions, whereas interactions in non-sandy media were dominated by favourable attractions to positively charged sites on clay edges and/or oxides as well as hydrophobic interactions with soil organic matter. The results may help to prioritize further research such as the currently unclear role of hydrophobic interactions in the fate of particles in porous media and identify the most important transport processes in more complex field situations.

## 1. Introduction

The use of nano-based technology in our societies has increased significantly in the last decades in the form of engineered nanomaterials (NMs). Some of these materials inevitably reach the environment and soils are no exception. It is for instance estimated that up to 44 % of the mass of some NMs having a $TiO_2$ or ZnO core are eventually emitted to soils (Kuenen et al., 2020). In addition, nanoplastics (NPs), often defined as plastic particles smaller than 1 μm, constitute a class of incidental, though anthropogenic, particles that is increasingly found in the environment including soils (Pérez-Reverón et al., 2023). An important pathway of both NMs and NPs into soils is via spreading of sewage sludge or other organic residues such as compost (Gottschalk and Nowack, 2011; Nizzetto et al., 2016; Weithmann et al., 2018). Some countries are now planning to ban use of sewage sludge on agricultural lands (e.g. Regerinskansliet, 2019), but there are several other pathways of NMs and NPs into soils. Unintentional emission may occur via wet or dry deposition (John et al., 2017). NPs have, for instance, been found in the most remote regions far from their atmospheric sources (Aves et al., 2022). Soils can also be exposed to anthropogenic particles emitted from traffic that results in road dust, which often contains NMs and NPs from worn tires, wearing off brake pads but also wear of pavement (Ermolin et al., 2017; Folens et al., 2018). Urban runoff also encompasses many sources of NMs of NPs from indirect emission during use, such as paintings from buildings (Kaegi et al., 2008), weathering of artificial grass (Horton et al., 2017) or plastic mulching which is applied directly to agricultural soils (Steinmetz et al., 2016). Finally, NMs or NPs can be intentionally emitted to soils via direct application, e.g. as (plastic-coated) nanofertilizers or nanopesticides, also referred to as nano-agrochemicals (Kah et al., 2012), or as remediation agents, the most used ones being different forms of nano zerovalent iron (nZVI) (Stefaniuk et al., 2016).

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[☆] This article is part of a Special issue entitled: 'Legacy in Nanosafety' published in NanoImpact.[☆☆] The current paper presents a meta-analysis of retention of nanomaterials and nanoplastics in porous media in an effort to identify the main retention mechanisms in porous media with an emphasis on natural media such as soils. This aligns very well with the aims and scope of Nanoimpact. The conclusions can contribute to refining environmental models of nanomaterials to also include transport of nanomaterials in soils where the transport fate predictor can be related to the most important soil properties.

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**Table 1**
Sourced studies sorted per nanomaterial or nanoplastic core material also showing the number of sourced observations of transport of each core material. Also shown are the Hamaker constants of these materials in water as literature source for these constants.

| Core material | # column tests | References column tests | $Log_{10}$ (Hamaker constant) (J) | Reference Hamaker constant |
|---|---|---|---|---|
| Ag | 102 | Braun et al. (2015); Cornelis et al. (2013); He et al. (2019); Liang et al. (2013a); Liang et al. (2013b); Lin et al. (2011); Mahdi et al. (2018); Rahmatpour et al. (2018); Sagee et al. (2012); Tian et al. (2010); Wang et al. (2014b) | −20.0 | Dagastine and Grieser (2004) |
| Alumoxane (R-Al-O) | 1 | Lecoanet et al. (2004) | −19.82 | Rosenholm et al. (2008)[a] |
| Biochar | 22 | Zhang et al. (2023b) | −19.46 | Xu et al. (2020)[b] |
| Cadmium selenide (CdSe) | 13 | Quevedo and Tufenkji (2012) | −18.96 | Bergström (1997) |
| Cadmium telluride (CdTe) | 17 | Quevedo and Tufenkji (2012) | −18.96 | Bergström (1997) |
| Singe walled carbon nanotubes (SWCNT) | 66 | Jaisi and Elimelech (2009); Jaisi et al. (2008); Lecoanet et al. (2004); Liu et al. (2009); Tian et al. (2010); Wang et al. (2014b) | −18.20 | Rajter et al. (2007) |
| Ceria ($CeO_2$) | 12 | Li et al. (2011) | −19.38 | Faure et al. (2013) |
| Copper (Cu) | 10 | Jones and Su (2012) | −18.76 | Bergström (1997) |
| Fullerenes ($nC_{60}$) | 79 | Lecoanet et al. (2004); Shen et al. (2015); Wang et al. (2014b); Wang et al. (2010); Wang et al. (2008); Zhang et al. (2012) | −19.11 | Ma et al. (2010) |
| Goethite (FeOOH) | 1 | Lecoanet et al. (2004), | −19.64 | Xu et al. (2015) |
| Gold (Au) | 38 | Norrfors et al. (2021) | −19.52 | Israelachvili (2011) |
| Hydroxyapatite ($Ca_{10}(PO_4)_6(OH)_2$) | 26 | Wang et al. (2012); Wang et al. (2014a); Wang et al. (2011) | −19.89 | Vandiver et al. (2006) |
| Magnetite ($Fe_3O_4$) | 6 | Ersenkal et al. (2011) | −19.37 | Faure et al. (2011) |
| Montmorrilonite ($(Na,Ca)_{0.3}(Al,Mg)_2Si_4O_{10}(OH)_2.nH_2O$) | 4 | Tian et al. (2010) | −20.14 | Weber and Kaufhold (2021) |
| Multiwalled carbon tubes (MWCNT) | 23 | Kasel et al. (2013a); Kasel et al. (2013b); Liu et al. (2009); Mattison et al. (2011) | −19.48 | Xia et al. (2019) |
| Polystyrene (PS) | 45 | Pelley and Tufenkji (2008); Quevedo and Tufenkji (2012) | −19.01 | Prieve and Russel (1988) |
| silica ($SiO_2$) | 7 | Vitorge et al. (2014) | −20.62 | Valmacco et al. (2016) |
| Silver sulphide ($Ag_2S$) | 7 | Norrfors et al. (2021) | −19.03 | Bennett et al. (1970) |
| Titania ($TiO_2$) | 5 | Solovitch et al. (2010); Sun et al. (2015); Wang et al. (2014b); Zhao et al. (2012) | −19.37 | Bergström (1997) |
| Zerovalent iron (nZVI) | 85 | He et al. (2009); Johnson et al. (2009); Jung et al. (2014); Laumann et al. (2014); Raychoudhury et al. (2014); Saleh et al. (2008) | −19.27 | Bergström (1997) |
| Zinc oxide (ZnO) | 24 | Sun et al. (2015); Zhao et al. (2012) | −19.72 | Bergström (1997) |

[a] An average value was taken for $Al_2O_3$ as a Hamaker for alumoxane could not be found.
[b] Average of Hamaker contants for 4 different biochar materials.

While there are other anthropogenic particles polluting soils, such as soot (Bird et al., 2015), NMs and NPs no doubt constitute the two most investigated classes of anthropogenic particles emitted into the environment as reflected by the swath of review papers summarizing aspects of their ecotoxicology and environmental exposure (Cornelis et al., 2014; Horton et al., 2017; Huo et al., 2022; Pérez-Reverón et al., 2023; Yusuf et al., 2024; Zhao et al., 2021). Most of these reviews focus on summarizing results obtained from ecotoxicity experiments conducted in soil microcosms that were homogeneously spiked with well-defined, newly produced, anthropogenic particles. Understanding exposure even in these highly idealised systems requires understanding particle fate given that particles will always have to migrate within the soil matrix towards biological receptors. Many of the aforementioned reviews therefore also focus on the fate of anthropogenic particles in soils by qualitatively reviewing the myriad of possible colloidal processes that can enhance or decrease transport distances of NMs or NPs. Such processes include homoaggregation of anthropogenic particles with themselves, heteroaggregation with natural suspended matter in soil pores, deposition on pore surfaces, the formation of an eco-corona, interaction with air-water interfaces (Cornelis et al., 2014).

Most of the knowledge on colloidal interactions of NMs and NPs has been obtained or inferred from a relatively rich dataset of transport experiments conducted using short, saturated, repacked soil columns. This dataset has, to date seldom been analysed quantitatively in a way that allows identifying the most important soil properties or colloidal processes amongst the many possible ones determining NM or NP mobility. Goldberg et al. (2015) and Goldberg et al. (2017) critically examined the quality of continuum models for the breakthrough curve and depth-dependent retention profiles obtained from a large number of column experiments, but did not focus so much on the effect of different soil properties on model parameters. Babakhani et al. (2017) were the first to use artificial neural network analysis to investigate how different continuum model parameters such as attachment ($k_{att}$) and detachment ($k_{det}$) rate constants fitted to a large number of column experiments are affected by different soil (or sandy media) properties. However, experimental $k_{att}$ values can be obtained in various ways, depending on the underlying model that is being used. Secondly, $k_{att}$ appear to vary to a large extent with experimental parameters such as column dimension and flow rate during the column experiment (Babakhani et al., 2017). Goldberg et al. (2015) therefore argue that more complicated models not necessarily provide more accurate parameters than simpler models.

Risk assessments of incidental exposures of soils to NMs or NPs or terrestrial applications of NMs and NPs would both benefit from at least a qualitative relationship between a fate descriptor of the mobility of these particles and soil properties highlighting the most important colloidal processes that steer NM or NP fate. The current paper presents a meta-analysis of experiments where NMs or NPs were injected in repacked, saturated columns. The global attachment efficiency ($\alpha_{global}$) was considered the parameter to best represent mobility of NMs and NPs in these columns. $\alpha_{global}$ is closely related to the mechanistic attachment efficiency ($\alpha$), a parameter that reflects the probability a NM or NP attaches upon collision with surfaces. However, when attachment efficiency, as in this study, is calculated from NM or NP recovery, the parameter is called $\alpha_{global}$, because surface attachment is in this case lumped together with other processes that affect retention of particles in the column such as detachment, hetero- and homoaggregation that increase the likelihood of straining (Hul et al., 2021). While $\alpha_{global}$ is, similarly to $k_{att}$, operationally defined to some degree (Norrfors et al.,

**Table 2**
Overview of metadata and the numeric range collected from 45 studies on the transport of NMs or NPs in short, saturated, repacked columns. For each predictor variable, the number of observations that reported this parameter is shown as well as the range and whether the variable was logarithmically transformed for principal component analysis and partial least squares analysis or in some cases whether it was not used at all during statistical analysis. Symbols and abbreviations are given wherever appropriate as well as comments on how metadata was collected and/or calculated.

| Parameter | # observations | Range | $Log_{10}$-transformed | Comment |
|---|---|---|---|---|
| *NM or NP properties* | | | | |
| Core material | 624 | See Table 1 | Not used* | Mineralogy of the core material was not considered. |
| Coating | 624 | See full database in SI | Not used* | The as-produced coating on the pristine NM or NP. |
| Diameter | 624 | 5–2758 nm | Yes | The hydrodynamic diameter measured in the feedstock suspension of the column test. |
| ζ potential | 584 | −114 – +32 mV | No | Calculated from the electrophoretic mobility measured in the feedstock suspension. |
| Hamaker constant | 624 | $2.4 \times 10^{-21}$–$6.2 \times 10^{-19}$ J | Yes | Sources: see Table 1 |
| Input feed stock mass concentration ($C_{stock}$) | 620 | 0.005–2486 mg $L^{-1}$ | Yes | |
| *Medium properties* | | | | |
| Texture (mass % sand, silt, clay) | 624[a] | Sand 5.9–100 %<br>Silt 0–78.7 %<br>Clay 0–47.6 % | Yes | There were 366 observations using a medium consisting of 100 % sand or glass beads |
| Total organic content (TOC) | 246 | 0–9.63 % | Yes | |
| Average aggregate diameter ($d_{50}$) | 624 | 0.04–1000 μm | Not used* | |
| Porosity ($\theta$) | 624 | 13–84 % | Not used* | If available, the effective porosity (instead of the total porosity) |
| Heterogeneity ($\Theta$) | 624 | 60.0–81.3 % | No | Calculated according to Babakhani et al. (2017) |
| Cationic exchange capacity (CEC) | 221 | 0.2–33.32 cmol $kg^{-1}$ | Yes | |
| ζ potential | 463 | −76.5 - +5 mV | No | The streaming potential measured in the mobile phase |
| *Experimental protocol* | | | | |
| Pore flow (approach) velocity | 624 | 0.003–167 m $d^{-1}$ | Yes | In many cases calculated from the experimental flow rate |
| Column length ($L$) | 624 | 27–300 mm | Not used* | |
| Column diameter | 624 | 6.6–120 mm | Yes | Inner diameter |
| *Mobile phase characteristics* | | | | |
| pH | 606 | 3–10 | No | |
| Concentration of mono- and divalent cations and anions | 497[b] | Monovalent cations: 0–125 mM<br>Divalent cations: 0–50 mM<br>Monovalent anions: 0–125 mM<br>Divalent anions: 0–2.3 mM | Yes | The concentration of all measured monovalent or divalent cations (e.g. $Na^+$, $K^+$ vs. $Ca^{2+}$, $Mg^{2+}$) or anions ($Cl^-$, $NO_3^-$ vs. $SO_4^{2-}$, $CO_3^{2-}$) was summed. A zero value means ion concentrations were not reported |
| Ionic strength (IS) | 608 | 0–150 mM | Yes | Calculated from the concentrations of mono/divalent cations/anions. A zero value means ion concentrations were not reported. |
| Dissolved organic carbon (DOC) concentration | 134 | 0–10′000 mg $L^{-1}$ | Yes | All concentrations of different dissolved organic materials were summed up. |
| *Experimental results* | | | | |
| Integrated eluted NM mass fraction ($R$) | 607 | 0–103 % | Not used* | Calculated as integrated eluted mass divided by feed stock mass concentration. |

* Several parameters were not used during partial linear squares (PLS) analysis because they were categorical (core material, coating) or they occurred in eq. (1) ($d_{50}$, $L$, $\theta$, $R$).

[a] Valid for sand % only.

[b] Valid for monovalent cations only.

2021), it is currently the only fate descriptor of NMs and NPs that at least conceptually bears potential to predict transport of particles beyond the column test it was determined in (Cornelis, 2015; Praetorius et al., 2014b), unlike $k_{att}$. Moreover, $\alpha_{global}$ is calculated without curve fitting from the experimentally observed mass retention of particles pumped into the column parameters and experimental parameters such as porosity, column length and flow rate using colloid filtration theory. This theory is thus the simplest possible model assuming one-site irreversible attachment only. While a simplification of reality in most cases, it allows for a uniform (re)calculation of $\alpha_{global}$ from experimentally observed recoveries of NMs and NPs across a large number of observations. The relationship between $\alpha_{global}$ and operational parameters such as column dimension or flow rate was analysed as well as the relationship with medium, particle and pore water characteristics. Variation of $\alpha_{global}$ with operational parameters ought to be limited to ensure that this parameter can be used outside the context of the column study it was determined in. In that case, $\alpha_{global}$ only varies with medium and pore water characteristics. Analysing the latter relationships allows determining what sort of colloidal processes are the most important ones steering NM or NP fate and how this varies with soil and pore water properties.

## 2. Material and methods

### 2.1. Data mining

A manuscript search was performed on Web of Science using the following keywords: ("soil" or "sand" or "porous media") and ("nanomaterial" or "nanoparticle" or "nanoplastic" or "carbon nanotube" or "fullerene" or "polystyrene") and ("transport" or "mobility"). From many possible NP polymer cores, only “polystyrene” was used specifically during data mining because of the relatively larger number of column studies that injected NPs of this polymer. Data of 624 saturated column tests reported in 45 manuscripts were thus sourced to construct a database of mobility experiments on NMs and NPs having a wide variety of core materials (Table 1).

The full database of metadata and breakthrough data of column tests is available in the Supporting Information (SI). While the resulting database does not include all possible transport studies done on NMs or NPs, the database includes a variety of porous media and core materials the properties of which cover the environmentally relevant ranges (Table 2). Most studies investigating transport of anthropogenic particles use glass beads or sand as these media have the highest hydraulic conductivity and also present the highest probability that breakthrough of NMs or NPs can be detected. The current study mined data both from studies using glass beads and pure sands as well as natural media such as soils but a preference was given to transport experiments conducted using natural media (Table 2 – Medium properties). In this way, the variance in medium (surface) properties was maximized allowing to investigate their relative importance in steering NM and NP fate. It may be argued that glass beads or homogeneous sandy media are not relevant for investigating the fate of NMs or NPs in soils, but these studies were included in data mining, because these studies often vary the chemistry of the mobile phase in terms of e.g. ionic strength (IS) and dissolved organic carbon (DOC) concentrations, whereas studies using natural soils tend to keep the chemistry of the mobile phase constant. Moreover, IS and DOC are not always measured during experiments with non-sandy natural media even though pore water chemistry in soils is highly variable as it changes with the medium (soil) properties. Including column studies using glass beads or homogeneous sandy media during data mining thus ensures that the effect of pore water chemistry variability on NM and NP fate is also captured. Moreover, natural sandy soils containing virtually no organic matter have a similar texture and porosity as the artificial sands and glass beads media used in some transport studies.

### 2.2. Attachment efficiency

The attachment efficiency ($\alpha$), i.e. the fraction of collisions between particles and surfaces resulting in attachment, is generally considered as the most appropriate fate descriptor for NMs in porous media. However, there is no generally agreed upon method to calculate this parameter from the results of column tests and $\alpha$ is calculated in many different ways. For instance, different convection dispersion equations can be used to derive different attachment rate constants from which $\alpha$ can supposedly be calculated. $\alpha$ is also frequently calculated using colloid filtration theory from the integrated recovery $R$ (integrated eluted concentration divided with integrated added concentration) of the NMs and NPs and other metadata using eq. (1).

$$\alpha_{global} = \frac{-2d_{50}}{3(1-\theta)L\eta_0} lnR \qquad (1)$$

Within eq. (1), $\alpha_{\text{global}}$ is the global attachment efficiency, $d_{50}$ is the average grain size of the medium, $\theta$ is the porosity and $\eta_0$ is the single-collector contact efficiency calculated according to Tufenkji and Elimelech (2004). Most studies that use eq. (1) do not differentiate between $\alpha$ and $\alpha_{\text{global}}$ terming both parameters as $\alpha$, but strictly speaking, this equation cannot be used to calculate $\alpha$. The recovery of particles ($R$) from column tests and thus also $\alpha_{\text{global}}$ vary with all processes that increase or decrease retention of particles in the column, whereas $\alpha$ is valid for only one type of particle interaction such as aggregation of two particles or attachment of one particle to a surface (Clavier et al., 2019; Hul et al., 2021).

Calculating $\eta_0$ requires Hamaker constants for which the values shown in Table 1 were used. Hamaker constants of a specific material reported in the literature vary widely depending e.g. on the measurement method used. Hamaker constants used in different column transport studies therefore also vary widely even if the same particle and/or medium is studied. The same value was therefore used consistently for the same material as shown in Table 1. The current study would benefit from a literature review aiming to obtain a consistent set of Hamaker constants that was measured and calculated using a consistent methodology. Such a study is unfortunately not available for all materials studied here and it was deemed beyond the scope of the current study to perform such a systematic analysis. Moreover, a natural soil is always a mixture of many different materials and a consistent methodology for estimating the Hamaker constant of such a complex mixture does not exist. In an effort to account for the effect of composition on the Hamaker constants of soils, these constants were calculated according to Cornelis et al. (2013) using the Hamaker constants in Table 1 and value of $5.1 \times 10^{-20}$ J for kaolinite (Weber and Kaufhold, 2021).

### 2.3. Heterogeneity

We calculated the heterogeneity of the columns according to the following equation taken from Babakhani et al. (2017) which provides a number between 60 and 90:

$$\Theta = 60 + 6 \times \left( \frac{1}{2} \times \frac{f_{clay}}{f_{silt} + f_{clay}} + \frac{1}{2} \times \frac{f_{clay}}{f_{clay} + f_{silt} + f_{sand}} + \frac{1}{2} \times \frac{f_{silt}}{f_{sand} + f_{silt}} + 3 \times \frac{f_{clay} + f_{silt}}{f_{clay} + f_{silt} + f_{sand}} + \frac{f_{clay} \times f_{sand}}{\left(f_{clay} + f_{silt}\right) \times \left(f_{sand} + f_{silt}\right)} \right) \qquad (2)$$

$f_{\text{sand}}$, $f_{\text{silt}}$,and $f_{\text{clay}}$ are the mass fractions of granulometric sand, silt and clay of the medium.

### 2.4. Regression analysis

Principal component analysis (PCA) was applied to explore covariance between predictor variables, identify outliers and observe clustering of experimental data. This was complemented with pairwise regression analysis between all predictor variables. Predictor variables such as column length and medium porosity were not included as these occur in eq. (1) and they thus have a direct linear relation with $\alpha_{\text{global}}$ values. Their direct effect on calculated $\alpha_{\text{global}}$ values would thus obscure the influence of other predictor variables during subsequent PLS analysis, which is essentially a linear model. It can be argued that the same is true for approach velocity and the Hamaker constant as these are also used to calculate $\eta_0$, but the relation between $\alpha_{\text{global}}$ values and these parameters is non-linear. Those variables that did not satisfy normality were logarithmically (base 10) transformed to reduce skewness, a commonly used approach (West, 2022). Moreover, in the case of PCA analysis, $z$ scores were calculated from transformed variables to ensure equal weight on the linear models. Regression analysis amongst predictor variables as well as PCA analysis were done using Matlab (v2020).

Partial least squares analysis (PLS) was then done to investigate which predictor variables were most important in explaining the variability in $\alpha_{\text{global}}$ values. The Matlab script used for this analysis is shown in SI. Logarithmic transformation and $z$ scores were also calculated for

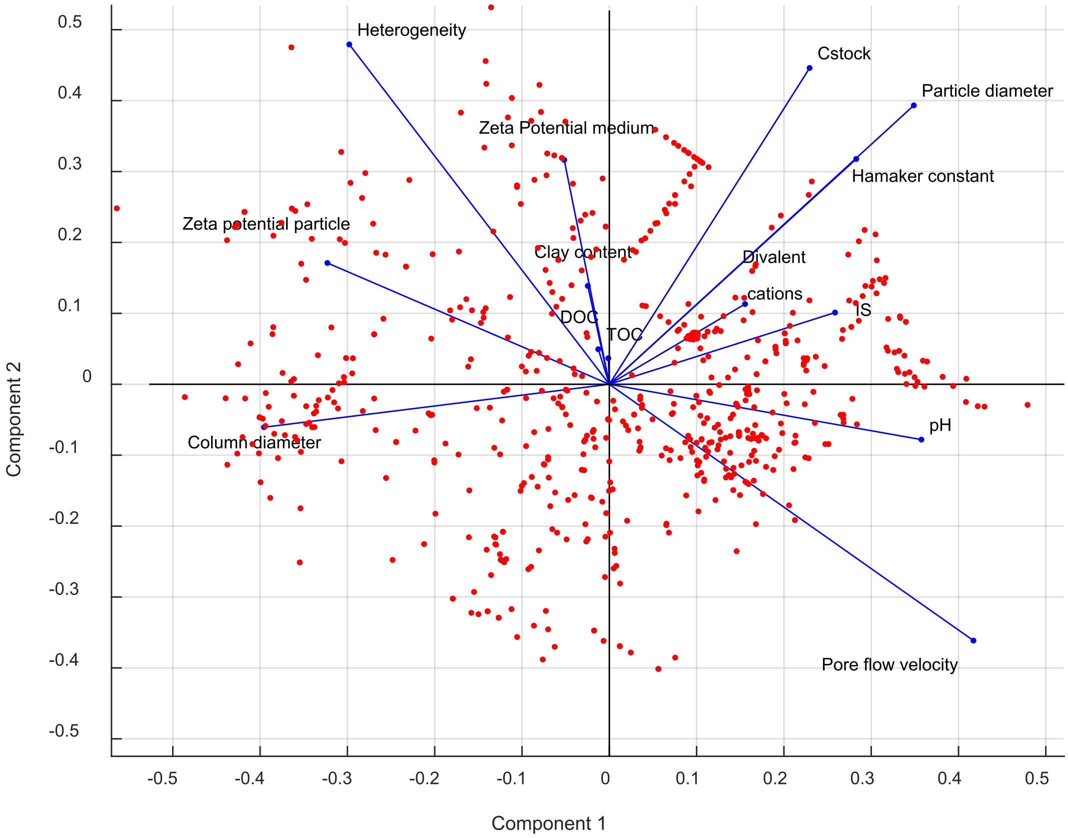

**Fig. 1.** Scores and loading biplot of the PCA-analysis.

the response variable $\alpha_{global}$. Logarithmically transformed $\alpha_{global}$ values are termed "$\log_{10}\alpha$ values" for the sake of brevity in the remainder of the manuscript. The SIMPLS algorithm was used to correlate $z$ scores of predictor values versus $z$ scores of $\log_{10}\alpha$ values. Normalising variables using $z$ scores is not required for PLS analysis per se. However, it was found that centring variables and reducing skewness had a high impact on the eventual ranking of importance of the variables by PLS analysis. There was, however, a high number of missing observations in several predictor variables or invalid results occurring after logarithmic transformation of zero values. The SIMPLS algorithm cannot take into account missing values in predictor variables which meant that full observations had to be discarded even if only one (or more) predictor variable had missing values. This severely limited the number of observations that could be used during PLS analysis if all predictor variables were to be considered at all times. Hence, predictor variables were progressively removed from PLS models, the ones with the highest number of missing values first, in an effort to better reveal how different predictor variables ranked in importance to explain the response variable. A PLS analysis was run in each case. The variable importance in projection (VIP) is a measure of the relative importance of different variables in determining the variability of a predicted value. The VIP of each variable was thus calculated using eq. (3) to reflect their importance in decreasing or increasing attachment of NMs or NPs in soils.

$$VIP_j = \sqrt{\frac{p\sum_{k=1}^{l}\left[SS_k\left(\frac{w_{jk}}{\|w_k\|}\right)^2\right]}{\sum_{k=1}^{l}SS_k}} \tag{3}$$

Here, $p$ is the number of predictor variables, $l$ is the number of latent variables. The number of latent variables was fixed at two, because two latent variables explained in all cases the majority of the variance in $\log_{10}\alpha$. $SS$ is the sum of squares relative to the $k^{th}$ latent variable calculated as $b_k^2 t_k^2$ where $b_k$ and $t_k$ are the predictor scores and response loadings, respectively. $w_{jk}$ is the weight of the j'th predictor variable, $||w_k||$ is the Euclidian norm of the weight vector of the $k^{th}$ latent variable. The VIP expresses the importance of each predictor variable in explaining the observed variance in the response variable. Usually, a VIP less than one signifies parameters that are less important and these predictor variables are usually omitted when building a PLS model used for prediction.

Finally, regression analysis of $\log_{10}\alpha$ with predictor variables was also done, using both log-transformed variables (according to Table 2) or $z$-scores. The slopes of the latter correlations better reflect the relative importance of correlations with $\log_{10}\alpha$ if found significant.

## 3. Results

### 3.1. Correlations amongst medium, mobile phase and particle variables

Table 2 shows how the different predictor variables were transformed to avoid skewed data. The granulometric clay content correlated highly with sand and silt content ($p < 0.001$). Hence, the predictor variable clay content was considered in further analysis, whereas sand and silt content were removed as predictor variables from further statistical analysis. Similarly, CEC and clay content were correlated significantly ($p < 0.001$). Geochemically, most cation exchange capacity is indeed generated by clays and oxides that mostly reside in the clay texture fraction explaining this correlation. However, CEC measurements are much more dependent on the specific experimental conditions under which they are measured compared to the clay fraction or the ζ potential of the medium. The CEC of soils was therefore not considered in further analyses.

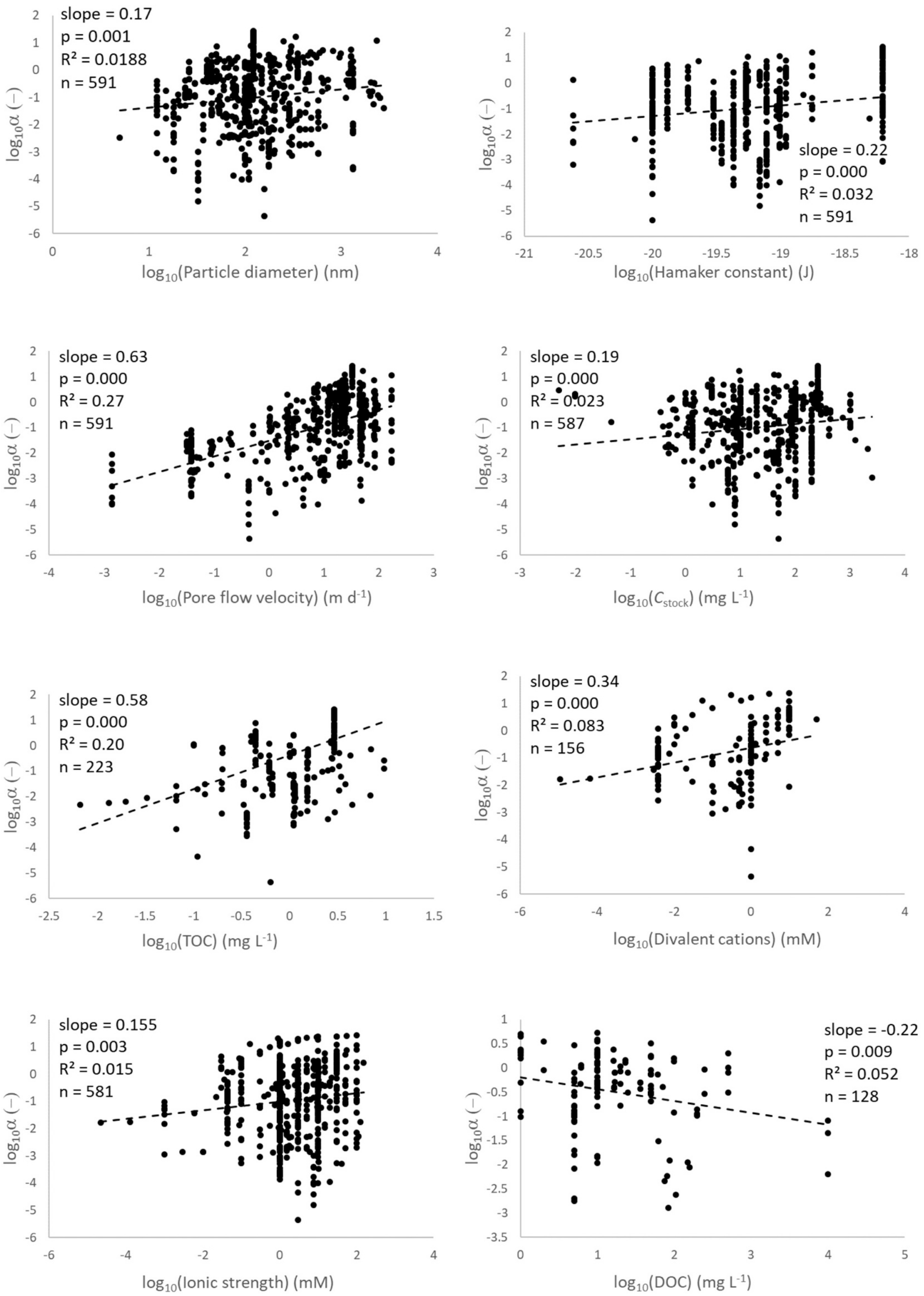

**Fig. 2.** Significant linear regressions of predictor variables with $\log_{10}\alpha$ for column tests taken across the whole dataset. The (normalised) slope, null hypothesis probability (p),Pearson correlation coefficients ($R^2$), and number of valid data points (n) are also shown.

A PCA analysis was done on *z*-scores of the remaining transformed predictor variables (Fig. 1) and all predictor variables were correlated against each other (See Supporting info, SI Table S1). Fig. 1 shows that no clear outliers could be identified during PCA analysis, but several correlations amongst predictor variables could be identified both during PCA analysis as well as during pairwise regression analysis. Not all of these correlations were relevant and some were likely purely accidental, such as the correlation between ζ potential of the particle and pore flow velocity. Other correlations were important to explain geochemical phenomena. The PCA analysis suggested non-zero concentrations of double charged cations (that could thus be log-transformed) correlated strongly with $\log_{10}$IS as confirmed in regression analysis (SI Table S1). However, divalent cations are more efficient than monovalent cations in shielding electrostatic repulsion between particles and surfaces. Divalent cations thus enhance NM or NP attachment much more efficiently compared to monovalent cations, a phenomenon called the Schulze-

Hardy effect. It was thus chosen to still include divalent cations in further analyses also given that PLS is relatively robust against multicollinearity. PCA analysis also suggested the TOC and clay content both correlated with the ζ potential of the medium, but the individual correlation with TOC was not significant ($p = 0.70$), whereas the one with the granulometric clay content was ($p = 0.001$). DOC and TOC were closely correlated and the particle's hydrodynamic diameter correlated significantly with the particle feedstock concentration and also, albeit less strongly, with the particle's ζ potential, the particle-medium Hamaker constant, pH, IS and the concentration of divalent cations ($p < 0.001$ in all cases). All these variables were included in the PLS analysis to probe the individual importance of these variables in inducing attachment of particles to soil surfaces.

### 3.2. PLS analysis of all column data

The full results of PLS analyses can be found in SI Tables S2, S3 and S4, whereas results of pairwise regression analysis of predictor variables with $\log_{10}\alpha$ are shown in SI Table S5. Note that pairwise regression models were always based on a larger number of observations than PLS models, because the former models require non-missing data for only one predictor variable, whereas the latter models require non-missing

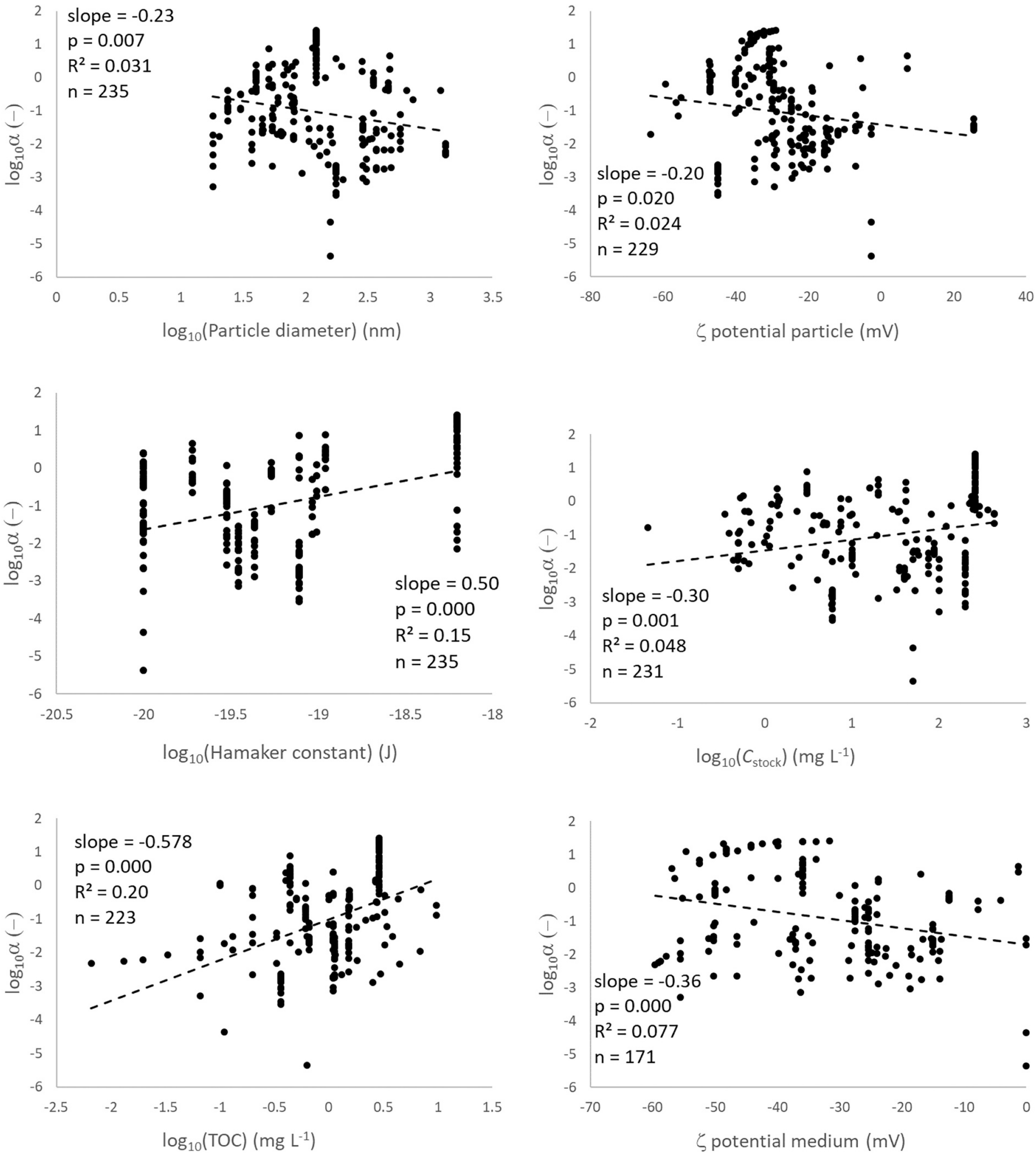

**Fig. 3.** Significant linear regressions of predictor variables with $\log_{10}\alpha$ for column tests for which the granulometric clay content of the medium was higher than zero. The (normalised) slope, null hypothesis probability (p), Pearson correlation coefficients ($R^2$), and number of valid data points (n) are also shown.

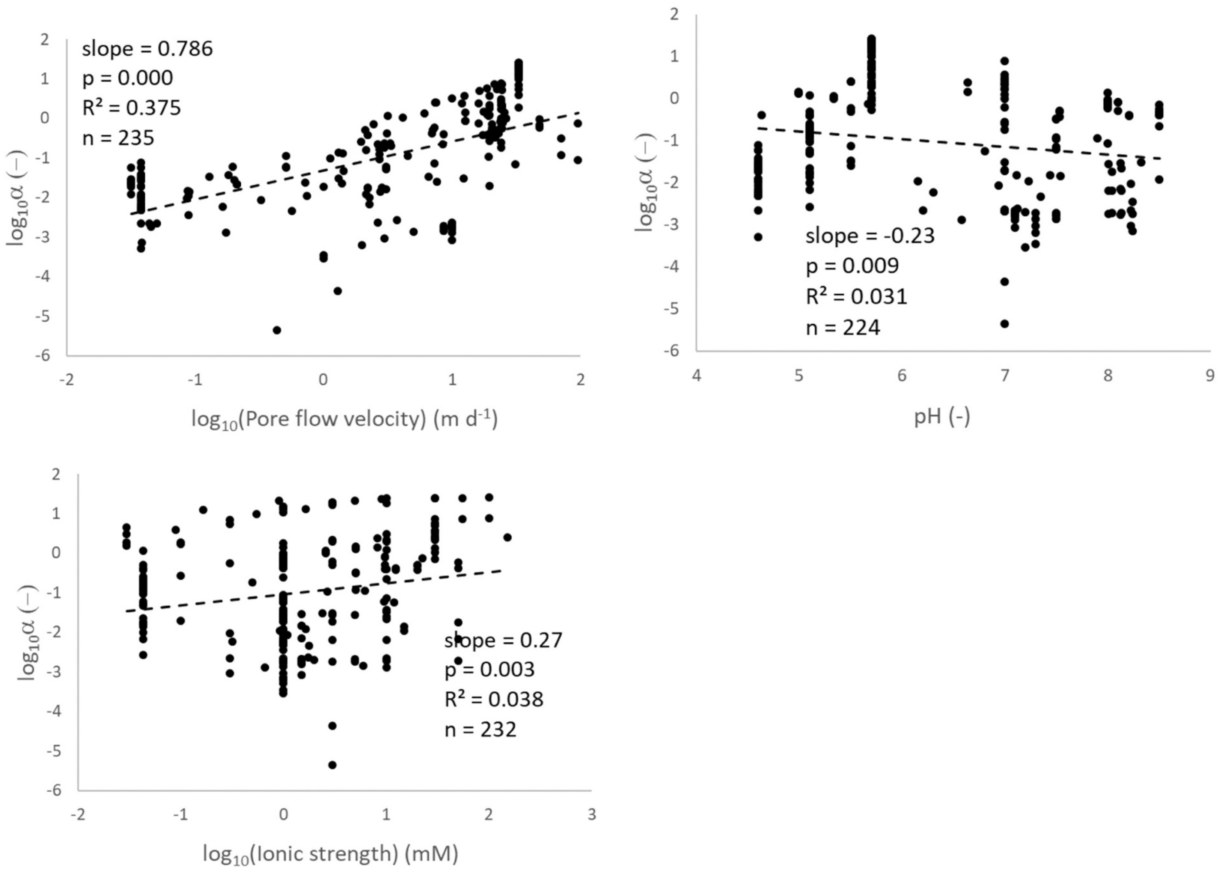

**Fig. 3.** (*continued*).

data for all included predictor variables. The first PLS model, i.e. using all predictor variables, thus only included 10 observations, because there were many missing or zero values for DOC and this parameter needed to be logarithmically transformed to satisfy normality. DOC is rarely measured in column studies using natural soils as a medium, but is sometimes added, e.g. in the form of humic or fulvic acids, in column studies using homogeneous sands as a medium. Formula (3) shows that VIP values are not normalised with respect to the size of the dataset. It is thus difficult to quantify the importance of DOC as a parameter or draw universal conclusions that generally apply to transport of NMs and NPs from PLS models that are based on only a few observations. Removing DOC as a predictor variable increased the number of observations in the PLS analysis from 10 to 76 and also increased the fraction of variance that could be explained by a two-component PLS model (from 55 % to 77 %). The number of included observations increased from 76 to 149 and then 157 when respectively the divalent cation concentration and TOC were removed as predictor variables from the PLS model, but the fraction of variance that could be explained by the PLS model was not altered much. Excluding the (granulometric) clay content from PLS analysis, however, increased the number of included observations from 157 to 399, but it also reduced the fraction of variance that could be explained by two components (from 78 % to 53 %) demonstrating the importance of this parameter. The clay content was also a parameter that had to be logarithmically transformed but a large number of observations (366 observations) stemmed from studies using pure sand columns where the clay content was zero. For this reason, separate PLS and regression analyses were also done for sandy columns, i.e. where the mass fraction of sand was at least 97 %, and for non-sandy columns, i.e. where the clay content was non-zero. A *t*-test revealed that average $\log_{10}\alpha$ values of both datasets were not significantly different ($p > 0.05$).

Not taking into account the first model that was based on only 10 observations, PLS done across the whole dataset indicated the Hamaker constant, TOC, ζ potential of the medium pH and in particular the pore flow velocity as important parameters explaining the variability in $\log_{10}\alpha$ values. These predictor variables attained VIP values above one in at least one PLS model. Linear regression analysis additionally indicated significant ($p < 0.05$) linear relationships between $\log_{10}\alpha$ values and the particle (hydrodynamic) diameter, $C_{\mathrm{stock}}$, the concentration of divalent cations, IS and DOC. The relationship with the ζ potential of the medium and pore water pH were, however, not found significant. Fig. 2 shows the significant relationships, which may help identifying whether the correlations are indeed important or e.g. leveraged by only a few observations. It can, for instance, be observed that the correlation with the Hamaker constants is leveraged by $\alpha_{\mathrm{global}}$ values obtained for experiments with single wall carbon nanotubes that had the highest Hamaker constant of $6.25 \times 10^{-19}$ as proposed by Rajter et al. A stronger conclusion with regards to the role of London and Van der Waals interactions could have been drawn if a set of Hamaker constants was available that had been consistently obtained and calculated with the same methodology for all medium and nanomaterials. Similarly, the relationship with IS and to some extent with the concentrations of divalent cations are leveraged by a few relatively low values. Moreover, some of the significant correlations having VIP scores lower than one (particle diameter, $C_{\mathrm{stock}}$, Divalent cation concentration, IS, DOC), are indeed weak having a low Pearson correlation coefficient and low normalised slopes (Fig. 2 and SI Tables 2 and 5). It is unclear whether the ζ potential of the medium and pore water pH are important variables explaining $\alpha_{\mathrm{global}}$ variability, because VIP values higher than one were attained for these variables in one PLS model but pairwise relation with $\log_{10}\alpha$ not significant. Variable selection via a combination of PLS analysis and calculation of VIP values is generally found superior to less advanced methods such as stepwise linear regression (Chong and Jun, 2005). However, VIP values higher than one were only attained in one PLS model based on only 76 observations for both parameters. The only parameters that appear as important in explaining the variability of $\alpha_{\text{global}}$ values of the whole dataset are thus TOC and the pore flow velocity, and possibly also the Hamaker constant that all have VIP values above 1 in at least one PLS model and show up as significant in pairwise regressions.

### *3.3. PLS and regression analysis of data from non-sandy media*

When only observations having non-zero clay content were analysed using PLS (SI Table S3), DOC was again the first variable to be excluded, because DOC is rarely measured in studies using non-sandy media. Model 1, i.e. where DOC was included, was thus valid for only 10 observations and strong conclusions can thus not be drawn from this model for reasons outlined earlier. 78 % of the variation in $\log_{10}\alpha$ was explained by the model where DOC was excluded and this fraction decreased as more and more variables were excluded in subsequent models. The VIP values varied somewhat over these models, but the pore flow velocity and the medium – particle Hamaker constant consistently

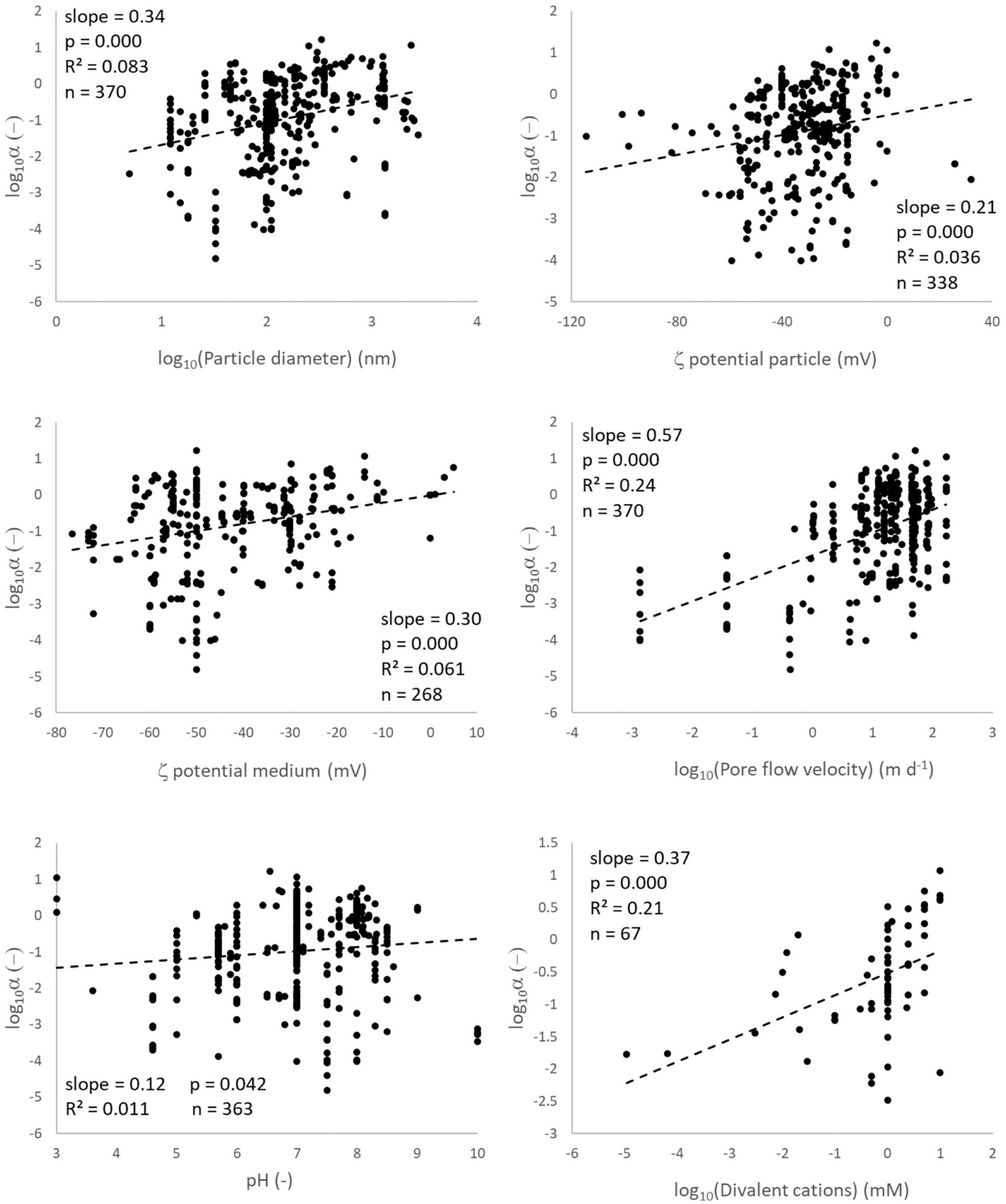

**Fig. 4.** Significant linear regressions of predictor variables with $\log_{10}\alpha$ for column tests for which the granulometric sand content of the medium was higher than 97 %. The (normalised) slope, null hypothesis probability (p), Pearson correlation coefficients ($R^2$), and number of valid data points (n) are also shown.

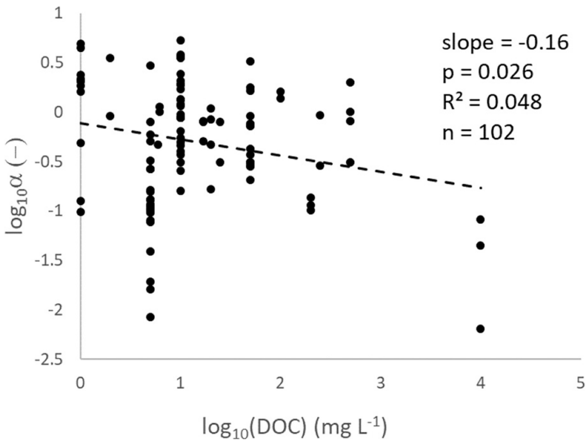

**Fig. 4.** (*continued*).

attained VIP values over 1. Other potentially important parameters that also attained at least one VIP value higher than 1 (excluding model 1) were TOC, ζ potential of the medium and pH. Direct regression analysis of $\log_{10}\alpha$ showed that these relationships were indeed significant (i.e. $p$ value $<0.05$, SI Table S5) and also the strongest ones having the highest normalised slopes. Several other significant relationships with particle diameter, ζ potential of the particle, $C_{\text{stock}}$ and IS were also found, but none of these predictor variables can be considered important to explain the variability of $\log_{10}\alpha$ values of non-sandy media. The VIP values of these variables were consistently lower than one and it can be observed in Fig. 3 how the pairwise relationship of $\log_{10}\alpha$ values with IS and the ζ potential of the particle are leveraged by only a few observations. Moreover, the significance and slope of the correlations with particle diameter and $C_{\text{stock}}$ are relatively weak.

### 3.4. *PLS analysis of data from sandy media*

The PLS analysis was repeated for observations where the granulometric sand content was at least 97 % (SI table S4). Again, successively fewer parameters were included and observations with at least one missing value in the remaining predictor variables were removed, thus successively increasing the number of observations. Note that clay content and TOC were never included in any model in this case as these variables were zero for most sandy media. The first two parameters to be removed were the concentration of divalent cations and DOC, an operation that increased the number of included observations from 8 to 70 and finally to 250. Many of the sand column experiments did not add divalent cations nor DOC to the mobile phase resulting in zero values for these variables that could not be logarithmically transformed. The first two models thus included a few, less than 70, observations. Moreover, they only explained a low fraction of the total variance of $\alpha_{\text{global}}$. A maximum of 57 % of the variance could be explained in the case of any PLS model for sandy media, i.e. a lower fraction compared to non-sandy media (78 %). PLS analysis and direct regression analysis were also less in agreement. Pore flow velocity again consistently attained VIP values above one, but also the particle diameter, ζ potential of the particle, the Hamaker constant, ζ potential of the medium, column diameter and pH emerged as potentially important variables explaining the variation of $\alpha_{\text{global}}$. Pairwise regression analysis with $\log_{10}\alpha$ did not indicate the correlation with the Hamaker constant nor with column diameter as significant (SI Table S5). In case of the Hamaker constant, VIP values higher than one were only attained in the first two PLS models valid for a subset of maximum 70 observations. The column diameter was conversely considered an important parameter, because it attained a VIP value higher than one in two PLS models, one of which was valid for 250 observations and explained 57 % of the variance in $\log_{10}\alpha$. In the case of the ζ potential of the medium a VIP value higher than one was only attained in the second PLS model valid for 70 observations. However, the significance and slope of the linear relationship between $\log_{10}\alpha$ and the ζ potential of the medium is relatively high, suggesting the latter parameter to be important. The relationships between $\log_{10}\alpha$ and respectively DOC and divalent cation concentrations were found significant (Fig. 4) even though these predictor variables did not attain VIP values higher than 1. However, PLS models that included these variables were only valid for a limited number of observations. Two additional PLS models were therefore calculated (SI Table S6) in which other parameters were omitted in an effort to include more observations with non-zero concentrations of divalent cations or DOC. The PLS model that included divalent cations was now based on 67 observations and a VIP value higher than one was calculated for divalent cations. The model including DOC was valid for 99 observations, but a VIP value lower than one was found for DOC. The particle diameter, ζ potential of the particle and medium, pore flow velocity, pH, divalent cation concentration and possibly column diameter were thus considered important parameters.

## 4. Discussion

### 4.1. $\alpha_{global}$ *values are to some extent operationally defined*

The strong effect of pore water velocity across the whole dataset and/or in individual sandy and/or non-sandy datasets confirms assertions that $\alpha_{\text{global}}$ values obtained from repacked column tests are to some degree operationally defined (Goldberg et al., 2017; Norrfors et al., 2021). This means that the fate of NMs or NPs cannot be accurately predicted in the field or in differently dimensioned porous columns using an $\alpha_{\text{global}}$ value even when that value was obtained in a column test using the same NM or NP and the same soil, but the column had different dimensions and/or a different pore flow velocity. The pore flow velocity is a parameter determining $\eta_0$ because a higher pore flow velocity tends to increase the number of collisions between particles and media grains (Elimelech et al., 1995). The correlation equation used to calculate $\eta_0$ has removed much of the variation owing to operational parameters, but not all of it. Using different correlation equations tends not to remove this type of variation completely either (Norrfors et al., 2021). The increasing (weak) effect of the feedstock concentration on $\alpha_{\text{global}}$ values (Fig. 3) has been observed before independent of which different correlation equation was used to calculate $\eta_0$ (Norrfors et al., 2021). This relationship can possibly be explained by a narrowing of pore necks by previously attached NMs or NPs, a mechanism that indeed becomes more important with an increasing $C_{\text{stock}}$ (Hul et al., 2021). A higher $C_{\text{stock}}$ also induces homo- or hetero aggregation and it is argued further that aggregates are more likely to be strained in pore necks.

$\alpha_{\text{global}}$ is, however, still the only parameter that can to any degree be related to chemical and mineralogical parameters of particles and soils to explain trends in particle mobility beyond column tests. Some adjustments to column test protocols may reduce the dependency of this parameter on operational parameters. For instance, $\alpha_{global}$ values tend to be determined using $C_{stock}$ and pore flow velocity values that are high compared to the situation in the field. Using these $\alpha_{global}$ values for field prediction would thus overestimate attachment and underestimate mobility of NMs or NPs. Using more realistic, i.e. low concentrations and low pore flow velocities during column tests could limit the named artefacts somewhat, but may also complicate analysis of breakthrough curves because of too low leached NM concentrations.

### *4.2. Homoaggregation occurs during column tests, but heteroaggregation is unlikely*

A higher $C_{stock}$ increases the probability that particles homoaggregate with each other in the feedstock solution or heteroaggregate with suspended solids in soil pores. Occurrence of homoaggregation in the feedstock suspensions was suggested by a significant increasing correlation between $C_{stock}$ and the particles' hydrodynamic diameter (SI Table S1). Moreover, the particles' hydrodynamic diameter was, as can be expected, significantly negatively correlated with the ζ potential of the particle and positively correlated with IS and the concentration of divalent cations in the feedstock. These correlations indicate how a reduction in repulsion between particles induces homoaggregation. Aggregation is often inhibited by formation of an eco-corona, but the negative relationship between hydrodynamic diameters and DOC in the feedstock was not significant. However, as argued earlier, homoaggregation during column tests is unwanted as this process is likely irrelevant for field situations in most cases, except when NMs are added intentionally to soils (Cornelis et al., 2014).

Extended heteroaggregation of NMs or NPs in pores e.g. with suspended solids, however, seems unlikely. Heteroaggregation of NMs or NPs occurs readily in pure suspensions of clays or oxides, especially if the NM or NP and clay or oxide surfaces have opposing charges (Cornelis et al., 2012; Hadri et al., 2018; Labille et al., 2015; Liang et al., 2013a; Qiu et al., 2020; Wang et al., 2015). Analysis of some soil column leachates with NMs reveals single NMs or small homoaggregates (Norrfors et al., 2021; Praetorius et al., 2017), but to the author's knowledge, no heteroaggregates have been observed in leachates of column tests with NMs or NPs. Some studies found heteroaggregates leaching from soil columns to which NMs were added, but only when the NMs were first attached to the pore wall from which colloids to which these NMs were attached were mobilized in a following step (Liang et al., 2013a; Makselon et al., 2017). Clays or oxides occur in soil pores of column tests because they were mobilized from pore surfaces owing to a relatively high surface potential (Norrfors et al., 2021) and/or an eco-corona (Praetorius et al., 2014a; Qiu et al., 2020) stabilising their presence in pore waters but also making heteroaggregation with NMs and NPs unlikely. Possibly, the hydrodynamic conditions in pore waters also benefit collisions between NMs or NPs and pore walls rather than with suspended solids, but this remains to be investigated.

While homoaggregation did occur, it likely had a limited effect on $\alpha_{global}$ values. This parameter lumps surface attachment together with other processes limiting particle transport such as homo- or hetero-aggregation and straining. Aggregation of nanomaterials reduces diffusion towards surfaces and thus lowers the attachment rate (Chen and Elimelech, 2006). Larger nanomaterial aggregates are thus expected to present comparably lower $\alpha_{global}$ values, but an opposite relation was found between $\alpha_{global}$ and particle diameter, except for the case of soil with a clay content higher than zero, where a weak negative relationship was found (Figs. 2–4).

### *4.3. Straining of single NMs or NPs or small aggregates unlikely*

It is often assumed that straining, i.e. physical filtering by pore necks or dead-ends, is an important retention mechanism of NMs and possibly also for NPs, especially if they first form homoaggregates (Yusuf et al., 2024). Larger particles or aggregates are indeed retained more as the probability of straining increases with the size of particles and straining is certainly relevant if NMs are intentionally added to soils in high concentrations such as during remediation of soils using uncoated nanozerovalent iron that tends to form large homoaggregates explaining limited transport distances in most porous media (Babakhani et al., 2018). At the same time, small single NMs and NPs are too small to experience any significant amount of straining. It is, however, less certain whether relatively small homoaggregates are retained by straining rather than by attachment to pore walls. The size-cut-off for straining to be relevant is often expressed in terms of the $d_p/d_{50}$ ratio where $d_p$ is the particle or aggregate diameter. The lowest such ratio is 0.002 as mentioned by Bradford et al. (2002), but this value was reported for relatively large ($> 2$ μm) colloids that individually had a relatively large probability of straining. The significant positive relationship between hydrodynamic diameters and $\log_{10}\alpha$ values for the whole dataset and for sandy media only seems to support that relatively larger aggregates are retained more, possibly by straining. However, an inverse relationship was found for non-sandy media and PLS analysis did not pick up particle diameter as an important variable promoting attachment. Homoaggregation of NMs and NPs thus appears to have a rather weak effect on retention. Straining may still play a role, however, at high $C_{stock}$ if a high number of pores is blocked by repeated attachment of NMs or NPs (Hul et al., 2021). Both, homoaggregation and pore blocking are, however, unlikely processes in the field where relatively low NP or NM concentrations occur unless added intentionally.

### *4.4. Attachment of NMs and NPs in soils is favourable, but unfavourable in sandy media*

A large number of studies emphasizes the importance of electrostatic repulsion in controlling the fate of NMs and NPs in soils. In an unfavourable attachment regime, the attachment efficiency increases if the negative surface potential of medium and particles are shielded more by higher background ion/cation concentrations. The positive relationships between $\log_{10}\alpha$, divalent cation concentrations and negative ζ potentials of particles or media indeed suggest an unfavourable attachment regime in the case of sandy media where different PLS models identified these relationships as important. Favourable attachment seems to conversely dominate in many of the non-sandy media where the effect of divalent cation concentration on $\alpha_{global}$ appear less important. NMs and NPs are small enough to interact with small localised patches of positive charge often found on clays or oxides even though the overall surface charge is negative (Alonso et al., 2009; Cornelis et al., 2011). Electrostatic attraction of particles by surfaces should more readily occur if more positive charges occur on the soil surface, which is the case at pH values lower than the points of zero charge of iron and aluminium oxides as well as clay edges, roughly occurring in a pH range of 6–8 (Sposito, 1998). $\alpha_{global}$ thus decreased with the medium's ζ potential becoming less negative (Fig. 3), which is not expected in an unfavourable attachment regime and a reverse relationship was found for sandy media. It thus seems that attachment in sandy media is mostly non-favourable, increasing with the concentration of divalent cations, whereas attachment is favourable in natural, non-sandy media in many cases and thus much less dependent on IS and more on soil composition (TOC) and to some extent on pH.

### *4.5. Soil organic matter may retain hydrophobic NMs and NPs*

Studying the role of natural organic matter in the fate of NMs is usually focused around the electrosteric stabilising effect of the eco-corona of NMs and NPs (e.g. Grillo et al., 2015). The significant positive correlation between $\log_{10}\alpha$ values and TOC, however, suggests that soil organic matter (SOM), i.e. static non-dissolved organic matter, may be an important attachment site of some NMs or NPs. NMs and NPs are

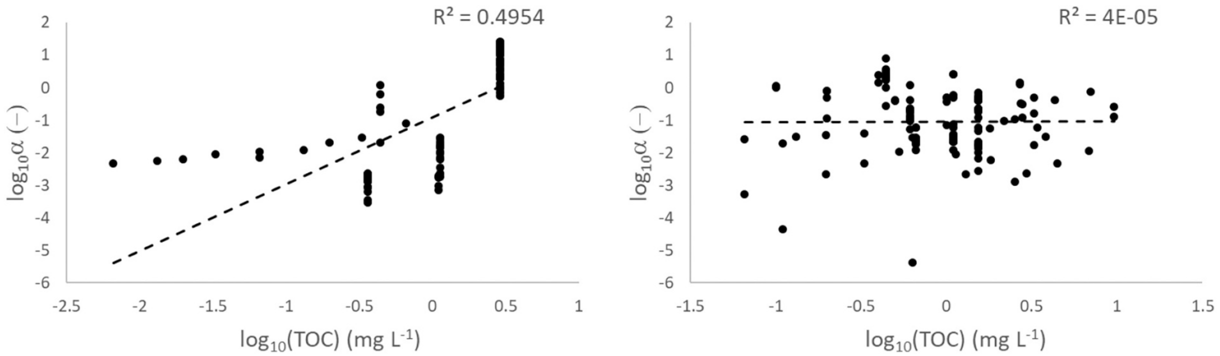

**Fig. 5.** Linear regressions of $\log_{10}\alpha$ against $\log_{10}$TOC for column tests where carbon-based particles (left) or non-carbon based particles (right) were eluted. The Pearson correlation coefficients are also shown.

repelled electrostatically by SOM which is predominantly negatively charged, but interaction with NMs or NPs may occur by other means. Only a few studies found a higher retention of NMs by organic rich soils and suggested hydrophobic interactions and/or cation assisted bridging flocculation may explain the attachment of NMs to organic matter (Adrian et al., 2019; Coutris et al., 2012; Mahdi et al., 2018). Cation assisted bridging may occur if a cation adsorbs to the negatively charged SOM causing charge reversal followed by attraction of a negatively charged eco-corona and/or the particle itself. There is no indication in the current dataset that this mechanism is relevant as $\log_{10}\alpha$ values were not very sensitive to an increase in the concentration of multivalent cations regardless of whether the TOC concentration was non-zero or not. The hydrophobicity of the particles in the current dataset likely varies widely as it contains data on both coated and uncoated inorganic and organic particles. Routine methods to characterise the surface hydrophobicity of particles have only recently been made available (Crandon et al., 2020; Valsesia et al., 2018) and hydrophobicity is rarely quantified in environmental experiments. It is, however, very likely that C-based NMs and NPs (NPs, fullerenes, biochar, carbon nanotubes, …) are on average more hydrophobic than non-carbon based NMs. The significance of the positive relationship between $\log_{10}\alpha$ and $\log_{10}$(TOC) exclusively for carbon-based respectively non-carbon-based particles, both shown in Fig. 5, were $< 0.001$ (103 observations) and 0.94 (126 observations), respectively. The difference in the trend of attachment between carbon and non-carbon-based particles supports that more hydrophobic particles indeed interact with SOM.

## 5. Conclusions

The presented meta-analysis highlights differences in attachment behaviour of NMs and NPs in various soil media, with key findings on electrostatic, steric, and hydrophobic interactions influencing retention and transport. In homogeneous, sandy media, several well-known colloidal processes such as electrostatic repulsion control the unfavourable attachment regime of NMs and NPs. Conversely, in more heterogeneous media such as natural soils, NPs and NMs likely attach favourably to clays and/or soil organic matter via electrostatic attraction and/or hydrophobic interactions. The latter process, however, warrants further research, especially in the case of NPs and carbon based NMs for which hydrophobic interactions may be particularly important. Column tests thus provide qualitative insights in NM and NP transport mechanisms and retention processes occurring in soils and column tests done with natural media such as soils seem most relevant for a field context where NMs and NPs are transported from topsoils. However, the quantitative applicability of parameters derived from column tests such as $\alpha_{\mathrm{global}}$ to predict transport distances in the field is limited, especially in unsaturated soils where air-water interfaces restrict transport (e.g. Kumahor et al., 2015; Zhang et al., 2023a) and in heterogeneous soils where macroporosity and/or non-advective transport processes such as bioturbation may both increase transport rates relatively to what is found in column test (Baccaro et al., 2019; Heinze et al., 2021; Heinze et al., 2024).

## CRediT authorship contribution statement

**Geert Cornelis:** Writing – review & editing, Writing – original draft, Project administration, Methodology, Investigation, Funding acquisition, Formal analysis, Data curation, Conceptualization.

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## Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

Geert Cornelis reports financial support, administrative support, article publishing charges, and travel were provided by Swedish University of Agricultural Sciences. Geert Cornelis reports a relationship with Swedish University of Agricultural Sciences that includes: employment. If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

## Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.impact.2025.100558.

## Data availability

All data is included in supporting info

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