RelationshipsbetweenBreakthroughCurvesandX-rayComputed TomographyAnalyzedMacroporeCharacteristics

TenundisturbedsoilmonolithsofclayeyPelosolatGottingen,Germanycoveringthehorizons$A_h$and$P$werecollected .Somecolumnswereleftatnaturalhumidity,somewereoven-driedtosimulatedrought situationsinforestsoilsinconsequenceofclimatechange.Insevencolumns,fourmicro-lysimeters,each, wereinstalledathalfheightinordertoobtaindataforanalysisofsinglesolutepathways.Afixedamount ofKBrtracerwasappliedtothehumuslayer.Thecolumnswereirrigatedwith$CaCl_2$.Columnoutputand lysimeteroutputwerecollectedandanalyzedtorecordbreakthroughcurves.Bimodalanalyticalconvection dispersion**e**quation(CDE)solutionswerefittedforthecolumnoutputsusinganon-linearleastsquarefit. AsimpleCDEsolutiondidnotfitwell.Thissupportsthemodeloftwooverlayingtransportphenomena. Afterbreakthroughrecordingwascomplete,allcolumnswerescannedusingX-raycomputedtomography (CT).FromtheCTdata3-Dreconstructionsoftheporoussystemwerecreatedforvisualinspection,and theexactpathwaysformacroporesalongthemicro-lysimetersweredetermined.Additionally,indicesof theporestructurewerecomputedtocomparewiththeslowandfastdispersivityvaluesfromthebimodal CDEfit.Thevariationinmicro-lysimeterperformancecouldbeexplainedusingthe3-Dreconstruction. Statisticallysignificantdifferencesbetweentheporestructureofwetanddriedcolumnsaftertheendof irrigationcouldnotbeidentified.Theporeindexhasgenerallyanegativelinearrelationshipwiththefast dispersivity,andapositivelinearrelationshipwiththeslowdispersivity.Theserelationsarestrongerin topsoil.TheCTpicturesand3-Dre-constructionsprovideaninterestinginsightintothesoilporesystem andmayhelptounderstandman-madedroughtproblemsduetoclimatechange.

Keywords: Tracertransport,X-raycomputedtomography,3-Dvisualization,bimodality,CDE,dispersivity, BTC/breakthroughcurves

Themainresearchquestionofthisworkwastofind out,ifsinglepathwaysinunsaturatedflowofwetand driedsoils(withregardtodroughtsduetoclimate change)couldbecharacterizedintheirfunctions. Quantitativerelationshipsbetweensoilstructure (especiallymacroporecharacteristics,namelytheirsize, number,type,distributionandcontinuity)and soilhydraulicpropertiesareessentialforimproving ourabilitytomodelflowandtransportinstructured soils.Bouma(1979)andothershaveusedchloride breakthroughcurvesfromundisturbedsoilcolumns asanindirectmeansofcharacterizingmacropores.In recentyears,X-raycomputertomography(CT)data, whichisbasedonvariedlinearattenuationofwater, airandsolidmaterials,providedanattractivetoolfor soilscientiststonon-invasivelyobservesoilstructure (GantzerandAnderson2002;Luo*etal*.2008;Taina *etal*.2008;Kumar*etal*.2010).Warner*etal*.(1989) usedCTdatatoinvestigatethemacroporesystem, *e.g.*characterizationofcracks,earthwormholesand rootingchannels.Peyton*etal*.(1992)concentrated onmacropores,quantifyingmacroporeperimeters,the pathsurroundingmacroporeshapes.Arelationship betweenmacroporesvisualizedbyCTdataand preferentialflowwasinvestigatedbyHeij*setal*. (1996).Luo*etal*.(2010)relatedmacropore characteristicsquantitativelytosolutetransport parametersundersaturatedconditions.Borgesand Pires(2012)havestudiedtherepresentative

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Table1. Description of experimental design

Column number	Treatment (type)	water content (%) (mean)	Br-Pulse t_0 [day] (Bram ount [mg])	Recovery rate (%)
1-4	Natural humidity (1)	37.5	0.833 (14.9-16.4)	76.9-85.9
5-7	Oven-dried (2)	30.7	0.833 (15.8-18.3)	55.2-91.1
10-12	Oven-dried (3)	30.7	0.0833 (15.3-17.3)	46.6-68.0

elementary area (REA) of soil samples using CT data, and concluded that samples with volumes from 50 cm^3 to 100 cm^3 with minimum cross section of 640 mm^2 are enough to be representative for the soil structure. Our soil samples had volumes of about 5000 cm^3 with a cross section of 169 cm^2.

The present study was carried out at University of Göttingen, Institute for Soil Science and Forest Nutrition, Büsgenweg, Germany on soil columns of a clay ey Pelosol (Vertic Cambisol) with well described swell-shrink characteristics (Spangenberg *et al.* 2011). The objectives were to study the role of pore contact of micro-lysimeters in the soil column on tracer transport in relation to the structure, as assessed by X-ray CT under different initial conditions and irrigation. As it is not possible to carry out these investigations under field conditions, non-destructive CT data were used, making it possible for 3-D reconstruction of the structure of the investigated soil columns. Furthermore, using CT data, an attempt was made to understand the relationship between the breakthrough curves of the tracers (velocity, dispersion and dispersivity) and the measured volume and structure of the pores.

Materials and Methods

The soil studied

The soil in the present study is a clay ey Pelosol (Vertic Cambisol), developed from a Muschelkalk (shell-lime) plateau derived from Triassic sediment layers. Collected horizons were $O_{l(f)}$–A_h–P, $O_{l(f)}$ was removed, only A_h–Premained. The P horizon showed a polyedric structure (loamy clay). In this horizon, swell-shrink characteristics are well expressed, and swelling clay minerals accounted for 26±5 %. Mineral compositions show quartz, illite, corrensite, orthoclase, albite and goethite. The details of the soil and the experimental setup were given in Spangenberg *et al.* (2011). A representative sample was taken in the area where the soil columns were taken. This sample was analyzed for soil density in various depths. Bulk density ranged from 0.908 Mg m^{-3} (topsoil, A_h) to 1.569 Mg m^{-3} (subsoil, P, 20-30 cm depth).

The column experiment

Originally, twelve undisturbed soil columns were collected in plexiglass cylinders of 14.7 cm diameter and about 30 cm height from a 4 m^2 sampling area. Ten columns were selected for this study, but in order to reduce the total amount of figures only columns 1, 5 and 12 are represented and described in detail. Table 1 shows an overview of the experimental design. Four columns (1-4) were kept at their natural mean water content of about 37.5% (mean value, n=4). Columns 5-7 and 10-12 were dried at 105°C in a heating cabinet to a water content of 30.7% (Table 1). Four micro-lysimeters, each, were installed in seven soil columns. These micro-lysimeters were equipped with a hydrophilic porous polymer tube (diameter 2.3 mm), made by Eijkelkamp Agrisearch Equipment, Netherlands (Spangenberg *et al.* 1997). They were designed specifically to minimize the impact of installation into soil. They were installed at about half the height of the column in order to allow observation of the flow parameters in higher temporal and spatial resolution in addition to the bottom output. All columns were irrigated in a vacuum packed plexiglas container, which is a complete soil microcosm (described in Spangenberg *et al.* 2011). Irrigation was applied using an electronic device every 2 h. At the bottom of each monolith, unsaturated flow was adjusted to 300 hPa. A bromide pulse was applied to each column at the beginning of the experiment. Then regular irrigation with $CaCl_2$ was started, which lasted for at least 30 days, *i.e.* until no more bromide was found in the column output. Solutions samples were collected daily at the bottom and from the micro-lysimeters of each column. They were analyzed for chloride and bromide. Analyses were done by HPLC ion chromatography. Elution mixture contained

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Na_2CO_3(2mmol)and$NaHCO_3$(0.75mmol). Detectionlimitofbothelementswas300µgL^{-1}.The statisticalanalysiswasdoneusingSAS(Statistical AnalysisSystem,SASInstitute,Inc.,Cary).The procedure"nlin"ofSASwasusedtoprovideleast squaresmodeling.Ananalyticalsolutionofthe convectiondispersionequation(CDE)wasusedtofit thebreakthroughcurvesoftheexperiment(vanGenucht en1982).Modelparameterswereestimatedusingbimodal ity,sinceitresultedinthebestfit (Spangenberg*etal*.2011).

BTCModeling

Themostimportantmodelforsolutiontransport insoildescribesconcentrationchangesintimeand spaceusingtheCDEaccordingtoNielsenandBiggar(1962).

$$R\frac{\partial C}{\partial t} = D\frac{\partial^2 C}{\partial x^2} - v\frac{\partial C}{\partial x} \quad \ldots(1)$$

where,*C*=concentration[ML^{-3}];*t*=time[days]; *x*=spatialdistance[L];*D*=dispersioncoefficient; *v*=averageporevelocity[LT^{-1}]

R=Retardation(dimensionless) $R = 1 + \frac{\rho_b K_d}{\theta}$ …(2)

where, ρ_b=soildensity[ML^{-3}];K_d=constantfactor[L^3M^{-1}];ρ =volumetricwatercontent[L^3L^{-3}]

AccordingtoToride*etal*.(1995)Rcanbeset on1.

Starting(1.0)andboundaryconditions(1.1and 1.2)are:

$$C(x,0) = C_i \quad \ldots(1.0)$$

$$\left(-D\frac{\partial C}{\partial x} + v\right)\Big|_{x=0} = \begin{cases} vC_0 & 0 < t \le t_0 \\ 0 & t > t_0 \end{cases} \quad \ldots(1.1)$$

$$\frac{\partial C}{\partial x}(\infty, t) = 0 \quad \ldots(1.2)$$

Assumingalteredinfiltrationanddisturbedwaterflo wduetosoildroughttheconditionsforapplication oftheCDEontreatmenttype2and3(Table1)were incomplete.Butconcentratingontheaimofthe investigation-acomparisonbetweendifferent experimentalvariantsofdriedandhumidsoils-the samemethodsofdataanalysishadtobechosen.To considerdifferentflowbehaviorofdriedandnon-driedsoilsomenewboundaryconditionsfortheuse ofananalyticalsolutionoftheCDEwereinvolved (NielsenandBiggar1962).Abimodalvariantofthis CDEwasused,moredetailsonthispartareprovided inSpangenberg*etal*.(2011).

X-raycomputedtomography

Aftertheirrigationphase,thecolumnswith installedmicro-lysimeterweretomographedina HospitalofGoettingenUniversity.AmedicalCTscanner (HiSpeedAdvantage,GeneralElectric)wasused.Forthep urposeofour30cmlongsoilcolumns, thehead-necksettingswerechosen.Tounderstand thedifferencesbetweenmoistanddrytreatments,each soilcolumnwastomographedindividually.Theoutput oftheCTunitisinHounsfieldUnits(HU),whichisanintern ationallystandardizednumberingscale (Petrovic*etal*.1982).Thenumericalvalueof HounsfieldUnitdependsontheattenuation coefficientsofthesubjectmatterrelativetothatof water(Hainsworth1983;Greverse*tal*.1989;Heij*set al*.1995),whichisgivenasH=1000(µ-μ_w)/(μ_w-μ_a). Here,µisthelinearattenuationcoefficientofthe materialorpixelin question,$\mu_w$and$\mu_a$arethe attenuationcoefficientsofwaterandair,respectively. A512by512matrixofpixeldatawasobtainedfor eachscan.Apixelhadthewidthofabout0.76mm. Thescanwastakenatintervalsof2mm(ca140 scanspercolumn).Aconstant1000wasaddedto everyvalue,whichisusedonallCTscanners,and giveninformulaforH.Afterthisaddition,airhasa valueof0andwater1000.Foreachpixel,theX-ray attenuationvalueswerestoredasvaluesfrom0to 4095.Thisrangewasduetothe12-bitprocessingof thetomographyequipment.Thehighvaluescorrespon dtothemetalofthemicro-lysimeter.The PV–WAVEwasusedforcomputeranalysis,which allowedreconstructing,visualizingandquantifying3-Dmacroporestructureinthesoilcolumn(Pierret*et al*.2002).

Indicatorsforporestructure

Themostimportantoutcomeofthe CTdataistheinfor mationonthecontentofsolidsandsoil porosity.The12-bitdatawasreducedto8-bitdata andthewholedatasetwasscaledbydividingallthe valuesbyafactor16,sothatwehavevaluesranging from0-255.Goodspatialresolutioncanbeachieved whenthereisalargedifferenceinHvaluesbetween asubject(*e.g.*asoilpore)andthebackground(*e.g.* soilmatrix)assuggestedbyGreverse*tal*.(1989).

Inthepresentstudy,thesameinteriorregionof thetop- andsubsoilinallthesoilcolumnswasselected,theareaaro undthemicro-lysimeterswasintentionallyleftout.Theselectedinteriorr egionsweredividedintoroughlycubicareas.Thesecubesf ormthebasisforfurtherdataprocessing.Idealcubes

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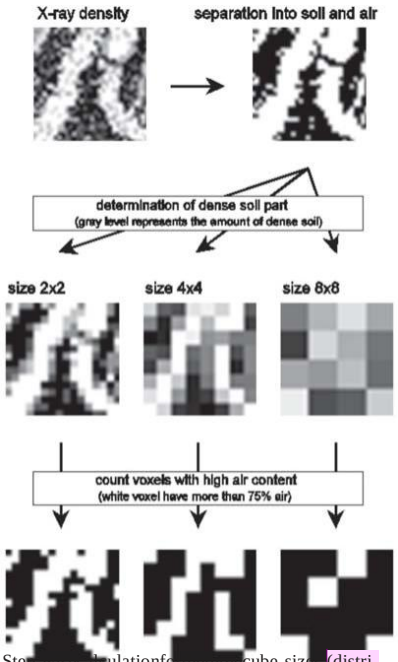

Fig.1. Step-wise calculation of the three cube-sizes. (distribution of X-ray attenuation values à separation of soil solids and air; determination of soil solids – (gray areas show the extent of soil solids); cube size 2×2 cube size 4×4 cube size 8×8 counting of cubes with high air volumes (white cubes have more than 75% air volume)

are not possible with the given voxel size. Therefore, different sizes were considered, namely, [4,4,1] voxel (labeled 2×2), [8,8,2] voxel (labeled 4×4) and [16,16, 4] voxel (labeled 8×8). These cubes correspond to volumes from 18 mm^3 to about 1 cm^3. The step-wise calculation for the 3 cubes is presented in figure 1.

First a threshold was selected to distinguish between dense matter and pores. A value of 110 was selected, as this provided the best separation based on known material locations. The percentage of cubes with less than 25% dense soil was computed for each sample area and cube size, which are labeled P_1, P_2 and P_4, depending on cube size. These were used as indexes for the porestructure of a column. For a completely homogeneous material, all indexes would be the same. We used P_1 as an index for the fine pore structure and P_4 is an index for the coarse pore structure. The indexes do not represent the connectivity of porous volume, only the amount of pores at a specific scale. The dispersivity values from bimodal tracer description were related to these indices for pore structure at three different scales. These indices were compared to the dispersivity results of the CDE models.

3-D reconstruction of soil structure

In the core of the column, which has a cross-sectional area of about 10cm×10cm, the voxels with scaled attenuation values from 0 to 30 were reconstructed as a 3-D image of soil structure. This allows a visual inspection of pore connectivity for qualitative analysis. A value of 30 was chosen to allow the air-filled inner part of macropores to be visible. For a better view of the pore contact of each lysimeter, the slice with the lysimeter was reconstructed separately in three viewing angles (40°, 70 and 85°) with same viewing angle in all cases, *i.e.* for all columns (Figs. 2, 3 and 4). However, only slice with 40 degrees viewangle was presented. In order to overcome the effect of air pockets in the outer space around the soil column, the central square area of 10cm×10cm was selected for the 3-dimensional reconstruction. At the top as well as at the bottom some slices of the column were discarded. The location of the micro-lysimeters in the column were established, in which the x-, y- and z-coordinates of the lysimeter head and terminal in the cross-section image were determined.

Results and Discussion

Influence of placement of micro-lysimeters and their function

The suction lysimeters of the tracer experiment drain different quantities of solution, although during the entire conduct of the experiment, a suction of 0.030 MPa was maintained. The great variability in the quantity of drained solution appears to be caused by the difference in positioning of the micro-lysimeters in the columns, relative to the porous system of the column. Preferential flow may initially have bypassed the lysimeters altogether, but relative position to the porous system seems to have a high influence. In order to explain this phenomenon, one column each group is presented in figures 5, 6 and 7. Each figure contains a cross section image at the height of the micro-lysimeters, a schematic of micro-lysimeter placement and the daily amounts of solution drained. The figures are used to judge the relative suction performance of the lysimeters. All four lysimeters of column 1 show similar performance (Fig.

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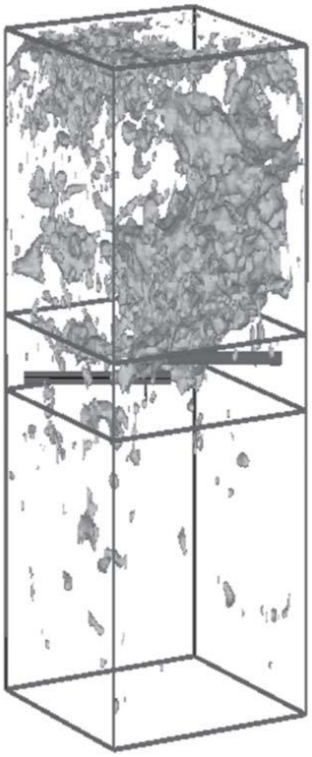

Fig.2a,Column1,withsection

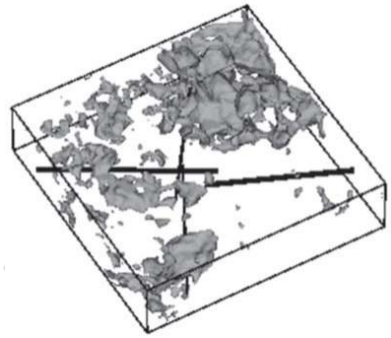

Fig.2b,Column1,section,40degree

Fig.2.3-Dreconstructionofcolumn1(experimentalvariant 1)withinstalledmicro-lysimeters.Inthecoreofthe column,thescaledattenuationvaluesfrom0to30are presented.Forabetterview,thesectionwiththelysim-etersispresentedina40degreelook-angle.Thecore ofthecolumnhasacross-sectionalareaof10cm×10cm

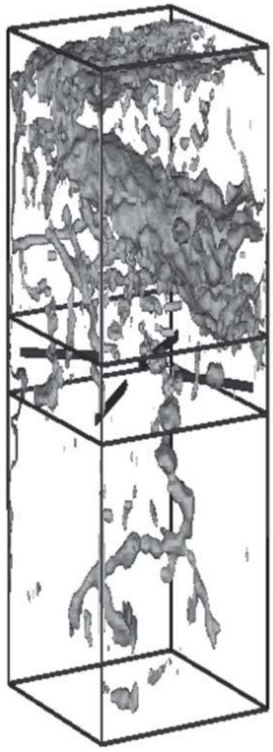

Fig.3a,Column5,withsection

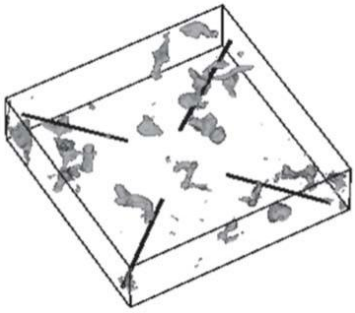

Fig.3b,Column5,section,40degree

Fig.3.3-Dreconstructionofcolumn5(experimentalvariant 2)withinstalledmicro-lysimeters.Inthecoreofthe columnthescaledattenuationvaluesfrom0to30are presented.Forabetterview,thesectionwiththelysim-etersispresentedina40degreelook-angle.Thecore ofthecolumnhasacross-sectionalareaof10cm×10cm

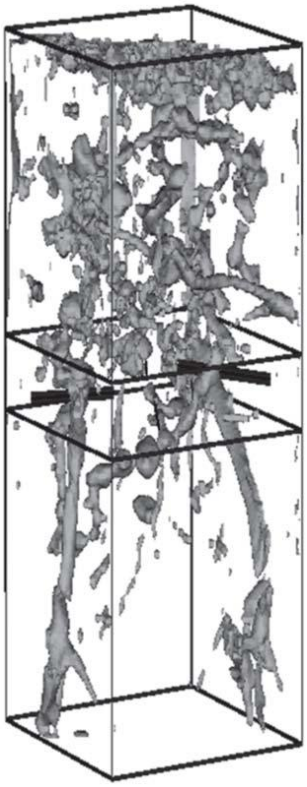

Fig.4a,Column12,withsection

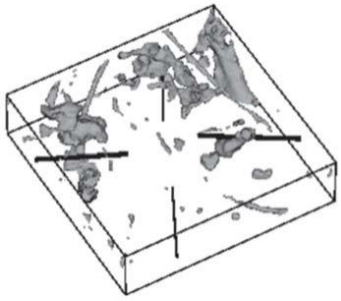

Fig.4b,Column12,section,40degree

Fig.4.3-Dreconstructionofcolumn12(experimentalvariant 3)withinstalledmicro-lysimeters.Inthecoreofthe columnthescaledattenuationvaluesfrom0to30are presented.Forabetterview,thesectionwiththelysim-etersispresentedina40degreelook-angle.Thecore ofthecolumnhasacross-sectionalareaof10cm×10cm

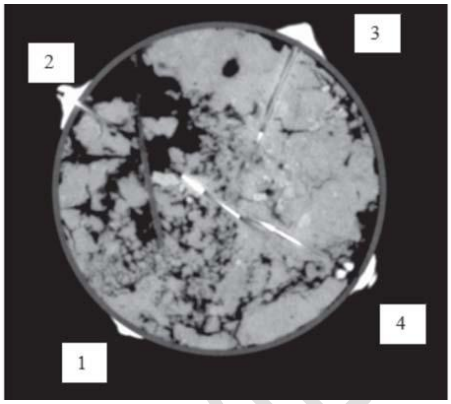

Figure5a,cross-sectionofcolumn1withmicro-lysimeters(topleft:no2;topright:no3;bottomleft:no1;bottomright:no4)andtheirnumbering

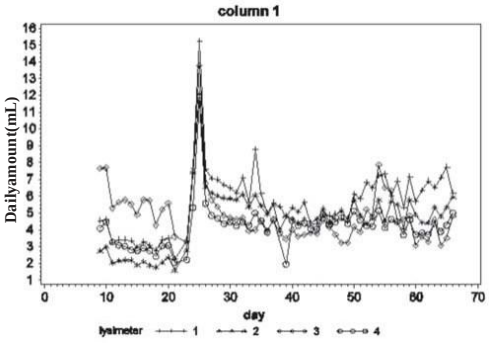

Fig.5bSamplevolumes(ml)ofcolumn1

Fig.5. Cross-sectionimageofcolumn1withmicro-lysimetersandtheirnumbering(5a)aswellasmicro-lysimeters'respectivemeasuredsamplevolumes(mL),column1(5b)

5c).Micro-lysimeters2and4showidenticalcurves, evenbeforeandafterthebreakincontinuityof leaching.Theirtipsliecloseto eachother(Fig.5a and5b),soweassume,thesesuctionlysimetersshowedr eciprocalinfluence.Modelingofthe breakthroughcurvesdidn'tmakesense.Incolumns5 and12,bothpreviouslydried,atleastonemicro-lysimeterhaddirectcontactwith a macrospore(Fig.6a, micro-lysimeterno4andFig.7amicro-lysimeterno 15).Thesemicro-lysimetersshowedthelowestsuction performancewithintheircolumn.Thus,contacttoa macropore(>0.5cm)appearstohaveaclear influenceonthesuctionperformanceofamicro-

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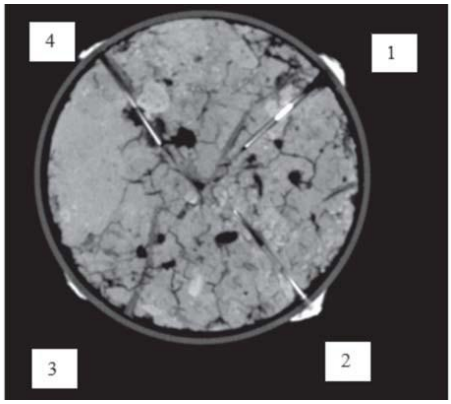

Fig.6across-sectionofcolumn5withmicro-lysimeters(top left:no4;topright:no1;bottomleft:no3;bottomright:no2) andtheirnumbering

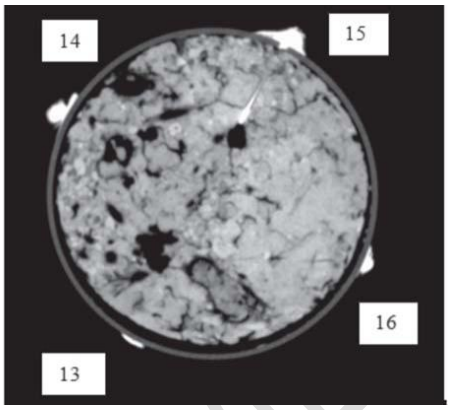

Fig.7aCross-sectionofcolumn12withmicro-lysimeters(top left:no14;topright:no15;bottomleft:no13;bottomright: no16)andtheirnumbering

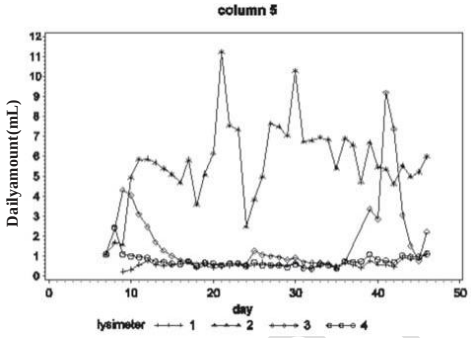

Fig.6bSamplevolumes(ml)ofcolumn5

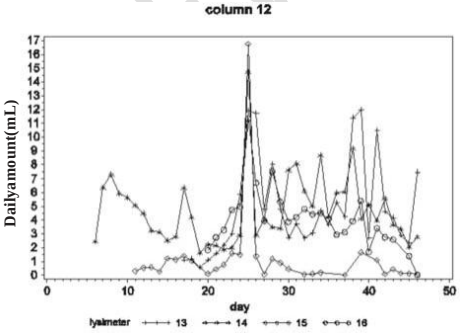

Fig.7bSamplevolumes(ml)ofcolumn12

Fig.6. Cross-sectionimageofcolumn5withmicro-lysimetersandtheirnumbering(6a)aswellasmicro-lysimeters'respectivemeasuredsamplevolumes(mL),column5(6b)

Fig.7. Cross-sectionimageofcolumn12withmicro-lysimetersandtheirnumbering(7a)aswellasmicro-lysimeters'respectivemeasuredsamplevolumes(mL),column5(7b)

lysimeter.Underunsaturatedconditions,onlyathin filmofwaterispresentonthewallsofporesofthis diameter.Becauseofthis,thesesuctionlysimetersdrained nosolutionmostofthetime,andhencelowvolumeofleachate.Transferringthisresulttonatural conditionswherethemicro-lysimeterswouldrepresent therootingzoneofforesttreesitcanbeassumedthat alreadyunderthesesimulateddroughtconditionswaters tresswouldhavebegun.

Micro-lysimetersinacompactsoilmatrixalso showedlowsuctionperformance.Aplacement avoidingtheseextremesresultedinsatisfactoryfunctioning.Ifenoughfineporesarepresentinthe vicinityofthemicro-lysimeters,thecapillary contributionofwateroutweighstheairfilledporosity, andthevolumeoftheleachatecollectedwillbehigher (Fig.8).Themediandailyamountofleachateafter theinitialphaseoftheexperimentisshownonthe abscissa.Ontheordinate,thecontentofthesolidsin thesoilinanellipsoidalregionaroundthetipofthe micro-lysimeter(ca.12cm^3)isshown.Interestingly, thesuctionperformanceofthemicro-lysimeterwithhighporevolumewasaspoorasforlysimeter swith almost100%compactsoilaroundit(column3).Most lysimetersshowbestperformancewithmoderately compactsoil.Inadditiontothetotalquantityofporesandsolidvolume,theporesizedistributionandtheir connectivitytotheimmediatevicinityofthelysimeter

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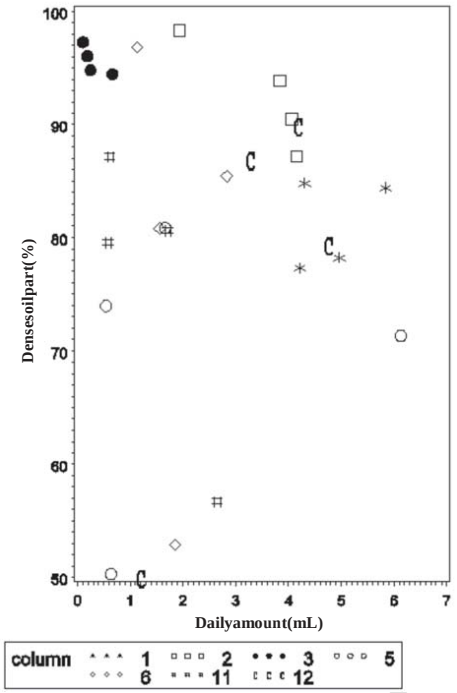

Fig.8. Percentage soil solids around the lysimeter in relation to the suction performance of the lysimeter for 28 lysimeters in 7 columns. The soil solid content was obtained from the CT attenuation data

is important. All these factors appear to be responsible for the otherwise unexplained suction effectiveness of the lysimeters. Eight of the 28 lysimeters have to be considered to be ineffective, since they drain less than 1 mL d^{-1} (Fig. 8). About 70% of the installed micro-lysimeters are capable of draining, and about half of them are distinctly useful (>3 mL d^{-1}) under the given conditions.

For the first time, CT analysis was able to visualize and reveal cause-effect relationships of micro-lysimeter pore contacts and their suctionability. The X-ray CT images of cross sections of the soil columns did not show noticeable disturbance in soil structure of the columns due to installation of the micro-lysimeters (Figs. 5a, 6a and 7a). In contrast to this, Beckmann *et al.* (1992) found noticeable disturbances in soil structure due to installation of standard lysimeter cups in the field, resulting in an alteration of the pore system. For most of the dried columns (columns 5 and 12; Figs. 10 and 11) the micro-lysimeters have missed the main breakthrough as they had no initial soil contact because of the dry state of the soil, or the first fast breakthrough bypassed them.

Localization of the micro-lysimeters and its relation to tracer breakthrough curves

Given that no CDE model could befit for the micro-lysimeter data, its breakthrough curves in relation to pore structure can only be discussed in a qualitative way. As examples the 3-D reconstructions of columns 1, 5 and 12 with installed micro-lysimeters are represented in figures 2, 3 and 4, respectively.

The breakthrough curves of all micro-lysimeters are available as well as all 3-D reconstructions of the scanned soil columns. The breakthroughs of the micro-lysimeters of column 1 (the wet variant of the experiment) along with the breakthrough of the whole column are represented (Fig. 9). In this column, the maximum values of the micro-lysimeter breakthrough curves were lower than the column output. Obviously, there it was a fast breakthrough, which bypassed the micro-lysimeters and a slow breakthrough that was eized by the lysimeter. The similarity of the micro-lysimeter breakthroughs is explained by the fact that all 4 micro-lysimeters lied close to and influenced each other (Figs. 2 and 5a). In columns 2 and 3 the micro-lysimeters, to a great extent, attain higher bromide values earlier than the column outflow

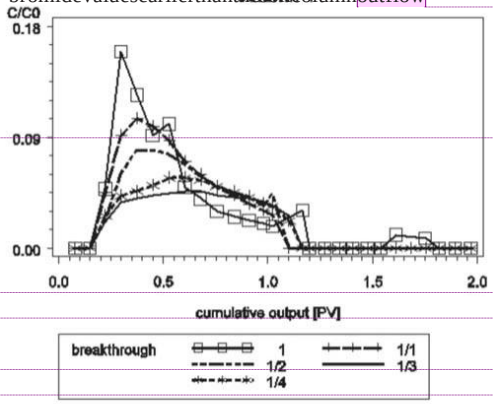

Fig.9. Breakthrough curves of column 1 (experimental variant 1) in units of the pore volume. C=measured concentration, Co=concentration of the Br pulses

Comment [D100]: *their*

Comment [D101]: examples,

Comment [D102]: 5,

Comment [D103]: 3,

Comment [D104]: available,

Comment [D105]: as are all

Comment [D106]: there was

Comment [D107]: through that bypassed

Comment [D90]: Percentage of soil

Comment [D91]: lysimeters

Comment [D92]: data.

Comment [D108]: outflow.

Comment [D93]: considered ineffective

Comment [D94]: cause-and-effect

Comment [D95]: between micro-lysimeter

Comment [D96]: in the soil

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Comment [D98]: 6a,

Comment [D99]: to the installation

Comment [D109]: concentration;

Comment [D110]: pulses.

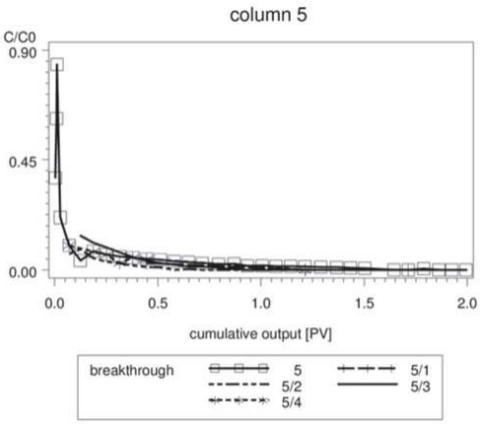

Fig.10. Breakthrough curves of column 5 (experimental variant 2) in units of the pore volume. C = measured concentration, Co = concentration of the Br pulses

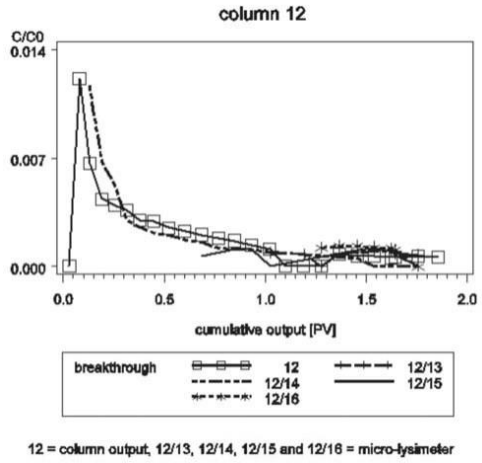

Fig.11. Breakthrough curves of column 12 (experimental variable 3) in units of the pore volume. C = measured concentrations Co = concentration of the Br-pulses

(not shown). These lysimeters have participated in the fast breakthrough via the coarse pores, *e.g.* column 2 (not shown, lysimeters 5 and 7).

At least one micro-lysimeter of column 5 (Figs. 3, 6a and 10) had very clear contact to the middle of a coarse pore, which traversed through the soil column, and which possibly contributed to the fast transport. Because of the deficient initial soil contact or too low suction capacity, this lysimeter did not show the fast breakthrough. A single lysimeter of the dry columns, lysimeter 14 of column 12 (Figs. 4, 7a and 11), showed an almost complete fast breakthrough. Of all the lysimeters of this column, this was the first to lead to soil solution leaching, and was present in the soil matrix with indirect contact to coarse pore space. This constellation, a mixing of indirect contact to coarse-pore transport space and direct contact to the fine-pore matrix, appears to have the best pre-condition for a quick functioning of the lysimeters, above all, in dried soil substance.

Pore connectivity in the columns and their importance for tracer transport

For a better understanding of the total transport through the columns, the transport into topsoil and in subsoil was considered. These two were characterized by a different soil structure and pore structure. The topsoil had a more or less strongly expressed crumb structure with high porosity, whereas, the porosity in the subsoil was low, because of the clay content. Hence, its structure was strongly influenced by roots, faunal burrows, and shrinkage cracks through drying, therefore, contains macropores. Both soil regions, according to their expression of their main characteristics, differently influenced tracer transport in a column.

Luo *et al.* (2010) investigated quantitative relationships between macropore characteristics and two major flow and transport parameters (K_{sat} and λ). Macropores played an important role. The traditional CDE modeled the BTCs well. Correlation between λ of the whole soil column and K_{sat} values of the B$_t$ horizon (not A) implied that the dispersivity was mainly controlled by the horizon with the lowest K_{sat} in the soil column. The most useful macropore parameters for predicting flow and transport under saturated conditions in the structured soils included macroporosity, number of paths, hydraulic radius and macropore angle. The presence of traversing pores (not necessarily macropores in the classical sense) in the subsoil influence the velocity and concentration of the tracers transport in this part. The reconstruction of the flow patterns is difficult, although the parameters are clearly defined. This is because of the interplay of all factors, with initial and boundary condition as water content can be different even in a small space but have a great influence. Further, the problem exists that only vertical transport mechanisms was considered. As is known from dye-experiments, the horizontal transport also spreads to a good extent (Ghodrati and Jury 1990), that is why, the vertically connected pores do not lead to preferential flow.

Comment [D111]: complete breakthrough.
Comment [D112]: lysimeters in this
Comment [D113]: contact with coarse
Comment [D114]: contact with the coarse
Comment [D115]: pore
Comment [D116]: contact with the fine
Comment [D117]: for the quick
Comment [D126]: *importance*
Comment [D127]: columns, transport
Comment [D128]: and subsoil
Comment [D118]: variable
Comment [D129]: drying; therefore, it
Comment [D130]: modelled
Comment [D131]: The correlation
Comment [D119]: concentrations
Comment [D132]: influences
Comment [D133]: tracer
Comment [D120]: *e.g.,* column
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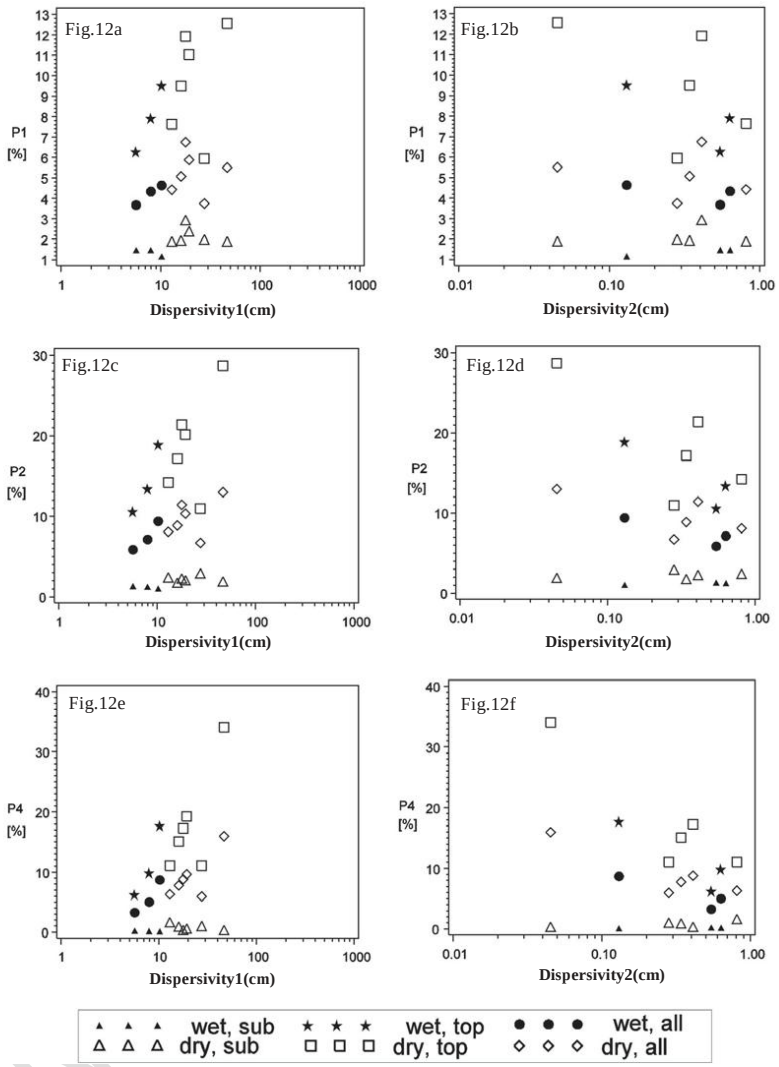

Fig.12. Dispersivity of the bimodal approach in relation to pore structure at three different scales (P1, P2, P4); three values for the respective column. Dispersivity 1 = D1/v1 = fast breakthrough, dispersivity 2 = *D2/v2* = slow breakthrough (Table 2). "Bottom" means subsoil of the column; "top" means topsoil and "comp." means complete column.

In the same way, Flury *et al.* (1994) reported that a major part of the water flowed past the soil matrix. As a result, an unexpectedly small portion of the soil, possible took part in the transport. Booltink and Bouma (1991) observed discontinuous macropores, so-called internal catchments, which should be integrated in water flow modeling. From studies on soil structure using x-ray CT, it was reported by Luo *et al.* (2008) that no macropores were continuous from the top to the bottom of the soil column, and some macropores became ineffective because of air-entrapment. They concluded that

Comment [D139]: P2, and P4

Comment [D140]: topsoil;

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Table2. Results of bimodal parameter evaluation. Retardation factor R=1, coefficient of dispersivity ($cm^2 Day^{-1}$), v=average pore water velocity ($cm Day^{-1}$), D_v/v_v=dispersivity (cm), Br=Bromide quantity of the impulses (mg).

Column	$D1/v1$ (cm)	$D2/v2$ (cm)	Br (mg)
1	9.0	0.18	14.3
2	8.0	0.63	16.0
3	5.7	0.54	15.0
4	14.5	0.3	15.7
5	19.3	0.0	32.4
6	38.1	0.08	34.5
8	16.0	0.34	14.3
10	24.4	0.49	16.4
11	23.1	0.26	16.8
12	12.9	0.81	11.5

macroporenetworkbyitselfcannotsimplybeequated toapreferentialflownetwork.Accordingly,inthe description offlow process,aseriesofcomplex processesneedtobeconsidered.

Inordertodescribetherelationshipbetweenthe BTCandthesoilporecharacteristics,we opposedfit parametersoftheCDEandsoilporeindices.As explainedearlier,theindexes(P_1,P_2andP_4)werecomputedf ortheporestructureofacolumn.P_1isan indexforporestructure insmaller scale.TheP_4on theotherhand,isanindexforcoarseporestructure (Fig.1).Therespectiveindices,P_1,P_2andP_4areshown ontheordinate,Ontheabscissa,eitherthedispersivityofthe slow(D_1/v_1)orfast(D_2/v_2) are shown(Fig.12, Table2).Thesymbolrepresentsthevariantofthe experiment(dry,wet)andtheareaofthecolumnthe indexappliesto(top=top-soil,bottom=sub-soil, comp.=completecolumn).

Infigures12b,12dand12f,correspondingfast dispersivityvalues(D_2/v_2)inrelationtoindexesforfine(P_1),middle(P_2),orcoarse(P_4)poreswere presented.Atallthethreescalesthesefiguresshowed anegativerelationshiptodispersivity,whichincreased withdecreasingaircontent.Thisrelationshipwas strongerforcoarserporeindexesintopsoil.The transportpathwayappeardtobeshortenedbyhigheraircon tent,whichloweredfastdispersivity.When coarseporeswerepresentinthecolumns(P_4),then withfastbreakthrough,uniformflowwastobe expected,withoutheterogeneityofpathway ofwaterandch emicalmovement.Nearlylinearlydecreasing linescouldbeobserved(Fig.12f)fortopsoiland wholecolumnindexes.Thishappenedtoalesserextentev enwithmorefinelydividedpore space (Fig.12d).Theslowdispersivityvalueshadapositive relationshiptotheporeindicesforallscales(Fig. 12a,12cand12e)andsoilareas,whichwasnearly linearinallscales.Withgrowingporecontentthe heterogeneityofthepathwaysincreased.Again,thisrelati onshipwasmoreapparent intopsoil.

Allresultshavetobeconsideredwithabitof caution,astheCDEfitwasusingamodel,whichfor thedrycolumnsmightbequestionable.Asa mathematicalmodelitreachedplausibledispersivityvalu es,though.Itexplainswhywecouldnotdetect differencesamongthesoilcolumnsjustby visualizing.Itcanbeassumedthatallcolumnsshowed macroporetransport,whichwasnotlinked to dispersivity ofthesub-soils.

Conclusions

The3Dreconstructionoftheporoussystemcan beusedtodiscussthesuctionperformanceof individualmicro-lysimetersandforexplainingthe breakthroughcurvesofmicro-lysimetersAnumeric poreindexrepresentingavolume'sporecontentataspecific scalecanbecomputedfromCTdata,and doeshavearelation toflowparameters.Thepore indexhasgenerallyanegativelenearrelationshipwith thefastdispersivity,andapositivelenearrelationship withtheslowdispersivity.Theserelationssuggest thatwithincreasingporosity,theheterogeneityofthe pathwaysincreases.WeconcludethattheCTimageslikecr oss-sectionsand3D re-constructions provide aninterestingandquiteuniqueinsightintothesoilporesyst emaftermoderatedrying.Cross-sectionsvisualizingtheporecontactofmicro-lysimeters'tipsmayhelptointerpretsoilwatermonitoringi nabetter way.

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Comment [D151]: In the topsoil

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Comment [D153]: subsoils.

Comment [D154]: to explain

Comment [D143]: of the flow

Comment [D144]: opposed the fit

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Comment [D156]: reconstructions,

Comment [D146]: is shown

Comment [D147]: At all three

Comment [D148]: appeared

Comment [D149]: of the pathway

Comment [D150]: space.

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