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Soil Infrastructure, Interfaces and Translocation Processes in Inner Space (“Soil-it-is”): towards a road map for the constraints and crossroads of soil architecture and biophysical processes

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Abstract

Soil functions and their impact on health, economy and the environment are evident at the macro scale but determined at the micro scale, based on interactions between soil micro-architecture and the transport and transformation processes occurring in the pore and particle networks and at their interfaces. Soil structure formation and its resilience to disturbance are highly dynamic features affected by management (energy input), moisture (matric potential), and solids composition and complexation (organic carbon, OC, and clay interactions). In this paper we review and put into perspective preliminary results of the newly started research program “Soil-it-is” on functional soil architecture. To identify and quantify biophysical constraints on soil structure changes and resilience, we claim that new paradigms are needed to better interpret processes and parameters measured at the bulk soil scale and their links to the seemingly chaotic soil inner space behavior at the micro scale (soil self-organization). As a first step, we revisit the soil matrix (solids phase) and pore system (water and air phases), constituting the complementary and interactive networks of soil infrastructure. For a field-pair with contrasting soil management, we suggest new ways of data analysis on measured soil-gas transport parameters at different moisture conditions to evaluate controls of soil matrix and pore network formation. Results imply that some soils form sponge-like pore networks (mostly healthy soils in terms of environmental functions), while other soils form pipe-like structures (poorly functioning soils), with the difference related to both complexation of organic matter and degradation of soil structure. The recently presented Dexter threshold (ratio of clay to organic carbon of 10 g g^{-1}) is found to be a promising constraint for a soil’s ability to maintain or regenerate functional structure. Next, we show the Dexter threshold may also apply to hydrological and physical-chemical interface phenomena including soil-water repellency and sorption of volatile organic vapors (gas-water-solids interfaces) as well as polycyclic aromatic hydrocarbons (water-solids interfaces). However, data for differently-managed soils imply that energy input, soil-moisture status, and vegetation (quality of eluded

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organic matter) may be equally important constraints together with the complexation and degradation of organic carbon in deciding functional soil architecture and interface processes. Finally, we envision a road map to soil inner space where we search for the main controls of particle and pore network changes and structure build-up and resilience at each crossroad of biophysical parameters, where, for example, complexation between organic matter and clay, and moisture-induced changes from hydrophilic to hydrophobic surface conditions can play a role. We hypothesize that each crossroad (e.g. between OC/clay ratio and matric potential) may initiate breakdown or activation of soil self-organization at a given time as affected by gradients in energy and moisture from soil use and climate. The road map may serve as inspiration for renewed and multi-disciplinary focus on functional soil architecture.

1 The Soil-it-is concept

The upper few meters of the soil sustain our life and society, but we only have an empirical knowledge on how soil behaves. As a contribution towards a better understanding of soil functional behaviour, we recently started an international framework program on functional soil architecture, called "Soil-it-is" (**S**oil Infrastructure, Interfaces and **T**ranslocation Processes in **I**nnner **S**pace).

With the Soil-it-is approach we wish to explore how soil architecture and infrastructure (pore and particle networks) control and are controlled by relatively poorly understood soil-physical and hydrological phenomena in all three soil phases (water, air, solids). The phenomena include soil-water repellency and fingered water flow, diffusive and convective gas transport in variably-saturated pore networks, and colloid mobilization/release and transport of colloids and colloid-bound chemicals.

The Soil-it-is vision is a holistic understanding of architecture and processes in soil inner space that will provide the basis for solutions to protect groundwater resources, improve clean-up technologies at polluted sites, enhance the health and productivity of cultivated soil, and improve the understanding of the Earth's critical zone (CZ), which

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is defined as the "heterogeneous, near surface environment in which complex interactions involving rock, soil, water, air, and living organisms regulate the natural habitat and determine the availability of life-sustaining resources" (NRC, 2001).

The soil vadose zone, being part of the CZ, is characterized by an immense activity of life, with microbes and fungi playing a key role in the organization and stability of soil organomineral complexes. These processes together have been labeled self-organization (Young and Crawford, 2004) and may be studied by reductionistic approaches at the level of microbes and isolated mineral particles. We aim at understanding and quantifying functional architecture and pore network dynamics as emergent properties at the aggregate to pedon scale. This is a prerequisite for, e.g., optimizing soil management in cultivated land productivity and soil recovery, prediction and control of soil greenhouse gas emissions and uptake, realistic risk assessment (chemical fluxes from soil to indoor air and to groundwater), and design of in-situ remediation strategies at urban polluted soil sites.

A platform hypothesis in the Soil-it-is project is that there is a lower limit (threshold) for soil organic matter (OM), below which it fails to support the self-organization and secure the functions and services described above. A low level of structurally active OM seems to dramatically increase the risk of clay dispersion. This in turn may increase soil internal vertical erosion. Mobilized clay colloids may be transported in macropores or inter-aggregate pores to the aquatic environments, strained at lower soil horizons, or crusted on the walls of soil inner space pores. To quantitatively evaluate this, gas transport studies provide an indirect but strong tool to describe the "morphology" per se of soil pores across scales, while mechanical disturbance of intact soil and fragments of soil in water in combination with analyses of structural strength across scales can be used to characterize the soil matrix per se.

In this study, we revisit recently measured data on different Danish soils supplemented by a few European soils. We re-analyze data to take a closer look at changes in functional soil architecture as derived from measurements of gas, water and solid-phase properties including diffusive and convective gas transport parameters, water

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dispersible colloids, soil water repellency, and sorption of dissolved and vapor-phase gases at different moisture (soil-water matric potential) conditions.

The study is divided into four parts. As a prelude, we argue for the need for a new and multidisciplinary research paradigm to understand and quantify functional soil architecture. Secondly, we take a renewed look at soil infrastructure with its complementary particle and pore network dynamics, hereunder suggest new ways of analyzing gas transport parameters measured at different moisture conditions to better reveal differences in functional network behavior and possible thresholds for self-organization. Thirdly, we consider a number of interface processes (water repellency and sorption of dissolved and vapor-phase chemicals) to see if the possible controls for soil self-organization and certain interface phenomena could be similar or related. Finally, we suggest a new concept for looking at functional soil architecture, making the first draft of a scientific road map to soil inner space where the crossroads between key biophysical parameters encounter gradients in applied mechanical energy and soil water intensity and hereby decides the dynamics of breakdown, resilience or build-up of soil micro-architecture. To navigate successfully and to find the mutual controls of soil structure formation as well as transport, translocation, and degradation processes will require a research effort comprising soil physics, chemistry, microbiology, and hydropedology with collaboration from several engineering disciplines. This is the ultimate goal of the “Soil-it-is” framework program.

2 Self-organization: the need for a new research paradigm

Science, and physics in particular, has developed out of the Newtonian paradigm of mechanics. In this world view, every phenomenon we observe can be reduced to a collection of atoms or particles, whose movement is governed by the deterministic laws of nature. Everything that exists now has already existed in some different arrangement in the past, and will continue to exist so in the future. In such a philosophy, there seems to be no place for novelty or creativity.

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Twentieth century science has slowly come to the conclusion that such a philosophy will never allow us to explain or model the complex world that surrounds us. Around the middle of the century, researchers from different backgrounds and disciplines started to study phenomena that seemed to be governed by inherent creativity, by the spontaneous appearance of novel structures or the autonomous adaptation to a changing environment. The different observations they made, and the concepts, methods and principles they developed, have slowly started to coalesce into a new approach, a science of self-organization (Heylighen, 2001).

Soil science has traditionally used reductionistic approaches, implying disruption of the subject prior to its investigation. Decades with this approach have improved our understanding of soil mechanisms (Six et al., 2004). Recent studies accentuate the continued need to uncover soil characteristics at the micro scale (e.g., Or et al., 2007; O'Donnell et al., 2007). However, soil is a very complex ecosystem, and the emergent functions and processes taking place in soil inner space cannot be interpreted from studies of isolated aggregates or micro-locations within the soil. Most soil functions and processes cannot be understood without acknowledging soil as a three-dimensional matrix (Letey, 1991; Karlen et al., 1990; Young and Crawford, 2004). Young and Crawford (2004) suggested that the soil-microbe system should be regarded as self-organized, i.e., that the organization will increase with time, without being controlled by environmental factors or an encompassing or otherwise external system. When a self-organized system is disturbed, its resistance to change from the equilibrium state as well as its resilience to a return to this equilibrium state is challenged (Holling, 1973). These classical concepts from ecosystem research extend our framework for interpretation of soil behaviour and help identify thresholds of soil disturbance that may be critical to a continued soil function (Carter, 2004).

Within the last two decades, a number of functional soil structure indices have been proposed that may both link soil structure to translocation and interface processes and help define the thresholds where soil structure is either improved, maintained (structure resilience) or destroyed in given processes. Examples include the Skopp

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et al. (1990) “Optimal Water Content” concept where solute and gas diffusivities are at a combined optimum for soil aeration and aerobic microbial activity, the Håkansson (1990) “Degree of Compactness” concept as linked to soil and crop productivity, the Moldrup et al. (2000) “Water-induced Linear Reduction” concept for separating resistances from solids and water on gas-filled pore network tortuosity and connectivity, the Or et al. (2007) “Soil Physical Constraints” concept emphasizing soil-water matric potential, water distribution and water film geometries and thickness as key controls of biological activity, biodiversity and bioactivity, and the Dexter et al. (2008) “Organic Carbon Threshold” concept separating clay-complexed or non-clay-complexed soil organic carbon (OC) in relation to soil functions and health.

Interestingly, as will be shown in this study, the various indices and concepts for functional soil structure (here called soil infrastructure) may be consistent with each other, both in terms of how key translocation parameters (e.g. gas transport parameters) can be used to characterize soil infrastructure and how parameter thresholds for soil structure (e.g. “clay saturation” by OC) control translocation and interface processes. To link the hitherto empirical soil infrastructure indices to a more conceptual understanding and quantification of soil architecture requires that many different physical, chemical, microbiological, and hydro-pedological parameters for both solid, water, and air phases and interfaces are measured on the same intact or minimally disturbed soil systems across moisture and management conditions. Some preliminary examples illustrating the usefulness of this research paradigm for understanding soil self-organization will be given in the next two sections on soil infrastructure and interface processes.

3 Infrastructure: the soil matrix and the pore system

3.1 The soil matrix

The key to soil self-organization is physical/chemical interactions between mineral and organic substances. These interact due to physicochemical processes. Dispersible

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clay and Fe- and Al-oxyhydroxides flocculate, influenced by organic bonding agents, e.g., extracellular polymeric substances excreted from plants, fauna, and microbes. At larger scales fungal hyphae and plant roots cross-link and enmesh the matrix. This traditional soil aggregation concept is described in Tisdall and Oades (1982) and references therein. Clay mineralogy governs the mechanisms active in aggregation, and most soils need OM to create stable macro-aggregates (Denef and Six, 2005). Recent achievements focus on the role of OM in re-arranging mineral particles into an open structure (Or et al., 2007). Clay minerals are the basic level in the hierarchy of soil structural elements. If the organization of clay particles – i.e. the interaction between clay and OM – is lost, all other hierarchical orders are lost or absent (Dexter, 1988). This may also be viewed as a collapse of soil self-organization. Re-orientation and hardening of dispersed clay minerals may result in a dense and mechanically strong but non-friable soil (Watts and Dexter, 1997; 1998). Evidence exists that OM in soil will reduce or prevent cementation (e.g. Utomo and Dexter, 1981; Kemper et al., 1987; Dexter, 1988). A similar situation exists in “hard-setting” soils of subtropical and tropical areas, where oxyhydroxides need OM to facilitate satisfactory tillage conditions (Mullins et al., 1987).

3.2 The Dexter (2008) threshold for complexed clay

Dexter et al. (2008) recently showed that arable soils often display a ratio, n , between $<2\ \mu\text{m}$ mineral particles (clay) and OC close to or higher than 10 ($n=\text{clay}/\text{OC}=10$). Their results further indicated that for such soils, some selected soil physical properties correlated to the content of OC. For mechanically undisturbed soils like pasture, they typically found clay/OC ratios lower than 10. Such soils are often described as having passed their “capacity factor” for carbon sequestration (Carter et al., 2003), and Dexter et al. (2008) found that the physical properties for these soils were not determined by OC but rather by the clay content. Dexter et al. (2008) defined “complexed clay”, CC,

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as

$$CC = \text{IF}[nOC < \text{clay}]\text{THEN}[nOC]\text{ELSE}[\text{clay}] \quad (1)$$

where $n=10$, and the amount of non-complexed clay, NCC, as

$$NCC = \text{IF}[(\text{clay} - CC) > 0]\text{THEN}[\text{clay} - CC]\text{ELSE}[0] \quad (2)$$

5 Dexter et al. (2008) showed that non-complexed clay is more easily dispersed in water than is clay complexed with OC. We have previously studied a range of arable soils (Schjønning et al., 2002a). For six of these soils having satisfactory tilth conditions the clay/OC ratio averaged ~ 9.7 . A similar calculation on data from the long-term fertilization experiment at Askov, Denmark, indicates that soil receiving either animal or
10 mineral fertilizer at adequate rates for a century in three replicate experimental fields had a clay/OC ratio of ~ 9.5 (Schjønning et al., 1994; Munkholm et al., 2002). According to the suggestion of Dexter et al. (2008), the clay of all these soils is virtually saturated with OC (clay/OC ~ 10). In contrast, soil unfertilized for a century at the Askov experiment had an average clay/OC ratio of 11.7. This is an indication of a pool of non-
15 complexed clay (NCC, Equation 2), and the soil treated in this way in all three replicate fields displayed severe signs of structural degradation (e.g., weak in wet conditions, mechanically strong in dry conditions). One of the soils studied in the Schjønning et al. (2002a) investigation was also depleted in OC due to long-term continuous growing of small-grain cereals without any return of organic residues and manure to the soil. For
20 that soil, the clay/OC ratio was as high as 13.7, and we found significant signs of degradation of soil structure (Munkholm et al., 2001; Schjønning et al., 2002a). A detailed study of water dispersibility of clay (WDC) by Elmholt et al. (2008) indicates, for soil samples collected across this Low-C soil – and only this soil – a correlation between WDC and OC expressed as the hot-water extractable C fraction. The above supports
25 the theory suggested by Dexter et al. (2008) of a lower threshold of OC for sustaining the self-organization process in soil. Their data actually revealed a clay/OC ratio for “saturation” of complexed OC on the clay particles in the range 8–11 (hence suggesting 10 as a suitable limit). This is in rather close agreement with the clay/OC ratios

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for the Danish arable soils reviewed above that have no tilth problems, while those displaying poor tilth conditions had higher values. More studies are needed, nevertheless, to investigate the general validity of one specific clay/OC ratio across soil types. The relation between clay and OC for the soils discussed here is shown in Fig. 1. We note
5 that the Dexter CC-NCC concept gives an explanation for the yet unresolved mystery that soils may exhibit satisfactory tilth conditions despite low OC contents as compared to structurally degraded soils with higher OC contents (e.g., the fertilized Askov soils compared to the Low-C of Group III).

3.3 Water-dispersible clay and aggregate stability

10 Several studies have shown that the water-dispersible clay (WDC) content is smaller when the soil OC content is larger (e.g. Watts and Dexter, 1997; Czyz et al., 2002). Dexter et al. (2008) found a higher correlation between WDC and soil clay content if NCC rather than total clay is used, which is in support of their hypothesis of two functionally different OC fractions. Based on our previous studies of the C-exhausted
15 soil mentioned above (Fig. 1: Low-C soil of Group III), we hypothesize that the self-organization of soils with $NCC > 0$ is threatened. Table 1 shows results from the Low-C soil and a neighbouring matching soil managed with forage crops and return of OM to the soil (High-C) (Munkholm et al., 2001; Schjønning et al., 2002a; Elmholt et al., 2008). We note (Table 1) that the soil content of total as well as a labile OC fraction
20 is significantly higher for the High-C soil than the Low-C soil. A calculation based on Dexter et al. (2008) found no NCC for the High-C soil, while the clay of the Low-C soil was not saturated ($NCC > 0$). In accordance with the general expectation, WDC of field-moist soil in a low-energy test was found to be significantly highest for the Low-C soil. However, it is noticeable that a significantly lower amount of WDC was found for
25 the Low-C soil when testing air-dried soil with the same energy input (Table 1). We interpret the results as an indication of cementation of readily dispersible clay in the drying process – and hence a decrease in dispersibility in a new short-term process of water application and energy input. This is supported by the high level of tensile

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strength of air-dried aggregates found for that soil; the cemented clay has formed internal crusts in the aggregates of the Low-C soil, hence producing mechanically stronger clods compared to the High-C soil. It is noteworthy that this is reflected also in a classical Yoder-type wet-sieving test; the Low-C soil displayed significantly *higher* aggregate stability than the High-C soil (Table 1). Other studies have likewise reported a poor correlation between dispersibility of clay and wet aggregate stability (Williams et al., 1966; Molohe et al., 1985; Pojasok and Kay, 1990). The effect of organic depletion on the soil matrix and soil pores is visually documented in Fig. 2.

Low levels of soil OM (NCC>0) may create aggregates that are unstable in wet conditions but become hard and strong in dry conditions (Watts and Dexter, 1997; Munkholm et al., 2001, 2002; Schjønning et al., 2002a). However, as discussed above, high stabilities of aggregates in wet conditions can be due to either cemented clay in low-friable clods or to biotic bonding and binding mechanisms in healthy crumbs (Schjønning et al., 2007; Elmholt et al., 2008). This has major impacts on agronomic aspects like tillage but also on clay dispersibility/colloid mobilization and hence colloid-facilitated transport of environmental pollutants (de Jonge et al., 2004a, b).

Colloid mobilization, stability, and transport in soil inner space are newly recognized and not-yet understood processes. The physical deposition of colloids on pore network walls (straining) may cause clogging of networks and dramatically changed soil architecture and infra-structure, affecting transport of colloid-bound chemicals through the soil vadose zone (de Jonge et al., 2004a; Sen and Khilar, 2006). These processes completely redefine our ideas about soil inner space dynamics. However, colloid transport has only recently been considered in models for transport and fate processes in the critical zone (Bradford and Torkzaban, 2008) due to a glaring lack of process knowledge. Soil physical disturbance, soil solution chemistry, and ambient soil moisture status all control colloid mobility (Kjaergaard et al., 2004) and thereby colloid-bound chemical transport (de Jonge et al., 2004a). Recent results further imply that the transport of colloids is controlled by mechanisms very different from those for dissolved chemicals and that soil structure (architecture) and preferential water flow are

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essential phenomena for quantifying colloidal transport in natural, undisturbed soils (Poulsen et al., 2006). Thus, there is considerable scope in studying colloid mobility and transport in the context of soil architecture and the self-organization concept.

3.4 Characterizing the soil pore system

Combined measurements of soil-gas diffusivity and air permeability are powerful means of characterising pore system dynamics in undisturbed soil (e.g., Ball, 1981; Moldrup et al., 2001). The breakdown of clay-OM interaction may induce distinct changes in the soil pore network. In a previous study (Schjønning et al., 2002b), we used a combination of gas diffusion and air permeability measurements to describe the pore system of intact soil cores collected in the same fields with contrasting management as addressed in the section on the soil matrix (Table 1). Figure 3 shows measured gas diffusivity for the two soils at four water potentials. The estimates of pore volumes blocked from the surrounding atmosphere (see data in the Figure) were found by assuming 1) a simple power law relationship between gas diffusivity and the air-filled pore space, and 2) a lower threshold of $D_p/D_0=10^{-4}$, reflecting no gas phase diffusion (Weast et al., 1983). We note that the High-C soil returns the highest estimate, which may be interpreted as a first indication of a more complex soil structure for that soil.

Moldrup et al. (2000) showed that for sieved, repacked soil in a wide texture range (between 6 and 54% clay) the soil relative gas diffusivity was well predicted by the model:

$$\frac{D_p}{D_0} = \frac{\varepsilon_a^{2.5}}{\Phi} \quad (3)$$

where D_p and D_0 are the gas diffusion coefficients in soil and air, respectively, ε_a is the volume of air-filled pore space, and Φ is soil total porosity. This so-called WLR (Marshall) model indicates that for soils with a non-complex soil structure, the reduction in relative diffusivity may be explained solely by a linear reduction of increasing water

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content (decreasing ε_a). We now hypothesize that the soil structure of the Low-C soil with dispersion-cementation cycles of clay – as discussed in the previous section – has degraded to mimic a sieved soil, i.e. obeying the WLR (Marshall) model for gas diffusion. In this exercise, we introduce the specific gas diffusivity as the ratio between the relative gas diffusivity and the volume of air-filled pore space:

$$\frac{D_p}{(D_0 \varepsilon_a)} = \frac{\frac{\varepsilon_a^{2.5}}{\Phi}}{\varepsilon_a} = \frac{\varepsilon_a^{1.5}}{\Phi} \quad (4)$$

Figure 4 (right) shows that for four matric potentials investigated, the specific gas diffusivity ($D_p/(D_0 \varepsilon_a)$) derived from measured data (black bars) is indeed approximately similar to those estimated from the WLR (Marshall) model for the measured ε_a values (grey bars). In contrast, the High-C soil interpreted as exhibiting an active self-organization (see the previous section) exhibits a specific gas diffusivity much lower than predicted from the WLR (Marshall) model. This reflects a much more tortuous/complex pore system than found for the Low-C soil. The same clear trend may be depicted from the independent measurements of convective gas flow in the pore systems. Groenevelt et al. (1984) suggested the specific air permeability as an index analogous to the specific gas diffusivity for expressing the continuity/non-complexity of soil pores. The specific air permeability is nearly an order of magnitude higher for the Low-C than for the High-C soil (Fig. 5). This means that the pore system of the Low-C soil is dominated by continuous (macro)pores enabling a fast convective transport of air through the soil.

Following Moldrup et al. (2001), but including the effect of inactive air-filled porosity (Brooks and Corey, 1966) as found in Fig. 3, we further evaluate the air-filled pore-network characteristics from a simple power-law model for relative gas diffusivity,

$$\frac{D_p}{D_0} = \left(\frac{(\varepsilon_a - \varepsilon_{ap})}{(\varepsilon_a^* - \varepsilon_{ap})} \right)^M \quad (5)$$

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where ε_{ap} is the inactive air-filled porosity or percolation threshold (Hunt, 2005), $(\varepsilon_a - \varepsilon_{ap})$ is the active air-filled porosity, M is a tortuosity-connectivity coefficient (Moldrup et al. 2001), and ε_a^* is a chosen reference air-filled porosity. We note that the often applied assumption of $\varepsilon_{ap} = 0.1 \times \Phi$ (Hunt, 2005) approximately applies to the High-C soil ($\varepsilon_{ap} = 0.051 \text{ m}^3 \text{ m}^{-3}$, $\Phi = 0.473 \text{ m}^3 \text{ m}^{-3}$), but largely over-estimates ε_{ap} for the Low-C soil ($\varepsilon_{ap} = 0.018 \text{ m}^3 \text{ m}^{-3}$, $\Phi = 0.420 \text{ m}^3 \text{ m}^{-3}$), suggesting much higher connectivity and continuity of inter-ped pores in the Low-C soil. We chose ε_{ap}^* at -100 hPa and note pores drained between -10 hPa and -100 hPa inter-ped or macropores (Moldrup et al., 2004) and pores additionally drained between -100 hPa and -300 hPa intra-ped or smaller pores. Figure 6 shows the plot of $\log(D_p/D_0)$ vs. $\log[(\varepsilon_{ap} - \varepsilon_{ap})/(\varepsilon_a^* - \varepsilon_{ap})]$ with the air-filled porosity at the reference point soil-water matric potential represented by the value of 0 on the x -axis (vertical dotted line). The slope to the left of $\log[(\varepsilon_a - \varepsilon_{ap})/(\varepsilon_a^* - \varepsilon_{ap})] = 0$ represents the inter-ped tortuosity-connectivity factor (M_1) and the slope to the right of $\log[(\varepsilon_a - \varepsilon_{ap})/(\varepsilon_a^* - \varepsilon_{ap})] = 0$ the intra-ped tortuosity-connectivity factor (M_2). The ratio of M_2 to M_1 ,

$$m = \frac{M_2}{M_1} \quad (6)$$

represents the change in pore network characteristics (connectivity, continuity and tortuosity) between intra- and inter-ped regions. The large m value of ~ 3 for the Low-C soil (as compared to ~ 1.5 for the High-C soil) implies a dramatic change in soil architecture between intra- and inter-ped regions, again supporting the hypothesis of a more pipe-like pore-network architecture for the Low-C soil. We therefore suggest m to be a useful soil architecture index.

Recalling the distinctly larger amount of non-complexed clay and hence the higher WDC for the Low-C compared to the High-C soil (Table 1), we note that the higher M_2 value of the Low-C soil (Fig. 6) may reflect internal crusting within peds for that soil. This interpretation needs to be confirmed by other studies, but has crucial impacts on several soil functions related to soil productivity as well as environmental aspects.

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3.5 Soil behavior at critical soil organic matter contents

Combining the pore system fingerprints obtained by the analyses performed above, we suggest that the pore system of a C-depleted soil may turn into a downpipe-like appearance in contrast to a well-managed soil exhibiting self-organization where our data indicate a “sponge-like” appearance (Fig. 7). We see a causal relation between the observed pore networks and the behaviour of the soil matrix when mechanically disturbed in wet and dry conditions. In Table 2, we summarize the characteristics of self-organized soils and soils where a low level of OM has inhibited self-organization. Dexter et al. (2008) found a linear relation between NCC and key physical soil properties. Our data indicate that non-linear behaviour (e.g. strongly cemented clods rather than friable aggregates) due to collapse of soil self-organization may be expected at considerable levels of NCC. It may be claimed that crusting and straining of clay particles is still an increase in organization. Thus Swift (1994) emphasized that resilient systems may have the capacity to occupy more than one state of equilibrium. There is, however, no doubt that a range of important soil functions in crop production and environmental protection will be poorer for the soil with a poor interaction between OM and clay particles.

4 Interfaces: processes in the solids-water-air continuum

4.1 Water repellency

Soil functions including water distribution and flow in the soil pore system are governed by interface processes. Water repellency (WR) is such an interface phenomenon, which affects hydrological processes or potentially all processes where water is involved. Critical WR can cause fingered flow (Fig. 8) and, in turn, preferential leaching of nutrients and chemicals to groundwater. At the smaller scale, the occurrence of interface-induced irregular water flow may also lead to altered soil inner space archi-

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ture due to accelerated particle dispersibility in the flow active architecture, which in turn further influences the flow pathways and vice versa.

The WR is closely related to soil-water content (de Jonge et al., 1999, 2007), OM quantity/quality (Kawamoto et al., 2007; Regalado et al., 2008), microbial activity (Hallett and Young, 1999), and soil management (de Jonge et al., 2007). Water repellency will at the same time control the onset of unstable (preferential or finger-like (Fig. 8)) water flow events in the soil (Nissen et al., 1999), thereby often causing a poor water distribution and accelerated transport of chemicals to the groundwater. Classical soil physics for describing water and chemical transport assumes that soil is non-water-repellent. However, recent evidence suggests that at least low levels of water repellency are the rule rather than the exception (Hallett and Young, 1999), and that spatial variations in soil OM quality and microbial activity control this so-called sub-critical water repellency. Consequently, small-scale changes in soil architecture including microbial activity and OM quantity and quality are likely major controls of flow heterogeneity and water distribution – where it was hitherto assumed that macropores and cracks in the topsoil were the main cause of preferential water flow and poor water distribution effects observed worldwide (Hallett et al., 2004; Or et al., 2007).

The WR varies non-linearly with soil-water content, w (King, 1981; Wallis et al., 1990; de Jonge et al., 1999, 2007; Kawamoto et al., 2007; Regalado et al., 2008) and all soils finally become wettable above a certain water content. This is illustrated (Fig. 9a) for a coarse-textured Danish soil from Jyndevad (Table 3) grown with three different crops of grass, barley and wheat. The trapezoidal integrated area, S_{WR} ($\text{mN m}^{-1} \text{kg kg}^{-1}$) under the WR- w curves (de Jonge et al., 2007; Regalado et al., 2008), can be used to determine the overall degree of WR for a specific soil (Fig. 9b). The grass crop clearly produces the highest WR followed by barley and wheat. The ways in which these soils differ are in the quantity and quality of soil organic C. With the same soil texture, what influences the overall level of WR is the type of crop grown on the soil and its interaction with the total amount of OC. The soils grown with small grain cereals have the smallest WR, but also a significantly lower content of OC (Fig. 9b). A comparison of the WR of

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soil collected from under a grass or barley crop shows a significantly higher WR for the grass crop despite the lower OC content of this soil.

Fractionating the Jynde vad soil collected from under grass crop allows us to investigate the effect of particles sizes on the degree of WR. The Jynde vad soil was fractionated into the fractions: <0.063 mm; 0.063–0.125 mm; 0.125–0.250 mm; 0.250–0.500 mm; 0.500–1.000 mm; and 1.000–2.000 mm. The degree of WR, S_{WR} , for the three smallest size-fractions (as well as the water-content at which the fractions becomes wettable) (Fig. 10a) increases with decreasing size-fractions (modified from de Jonge et al., 1999). Taking another and a closer look (Fig. 10b) by plotting the S_{WR} for all fractions as well as the whole soil as a function of soil OC reveals that the degree of WR is actually related to the OC content in the fractions rather than the particle sizes themselves. The OC content obviously plays a dominant role for the water repellency interface phenomenon. In the literature significant effects of OC on a range of soil physical properties have been reported including compactability, tillage, stability in water, and soil physical quality in general (Dexter et al., 2008 and references therein). Usually the effects of OM are such that an increased OM content results in improved soil physical properties. But as for the effects of increased WR with increasing OC content and the possible resulting irregular water movement the positive role is less clear.

4.2 The Dexter (2008) threshold for complexed organic carbon

In line with the complexed clay concept described in Sect. 3, Dexter et al. (2008) introduced the term complexed organic carbon (COC). This involves the idea that 1 g of OC is associated or complexed with n grams of clay ($n=10$). The amount of COC can be calculated by

$$\text{COC} = \text{IF}[\text{OC} < \text{clay}/n]\text{THEN}[\text{OC}]\text{ELSE}[\text{clay}/n] \quad (7)$$

Likewise the amount of non-complexed OC (NCOC) can be calculated by

$$\text{NCOC} = \text{IF}[(\text{OC} - \text{COC}) > 0]\text{THEN}[\text{OC} - \text{COC}]\text{ELSE}[0] \quad (8)$$

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The OC contents of numerous Jynde vad soil samples (de Jonge et al., 2007) having undergone different treatments are plotted as a function of their clay content (Fig. 11a) along with a number of other soil types, for which the WR-w curve has been determined (de Jonge et al., 1999). All of these soils fall well above the saturation line, which implies that they hold smaller or larger amounts of OC in a non-complexed form (NCOC). The degree of WR for these soils, as determined by S_{WR} , plotted against the amount of NCOC yields a linear relationship with a Pearson correlation coefficient of $r=0.77^{***}$ (note the data points from the Jynde vad grass field, the upper open circles, have been omitted in the correlation analysis due to the influence of eluded OC from the crop). This is only a slight improvement as opposed to correlating against OC ($r=0.76^{***}$, analysis not shown), but the regression line hits the origin when correlating with NCOC (interception not significantly different from zero), which is not the case when using OC (analysis not shown). This points towards the WR phenomenon being switched off when no NCOC is present in the soil.

The above analyses imply that OC quantity and quality, soil texture (specific surface area) and soil structure including aggregation may all in combination with actual soil moisture conditions (content and distribution) play a key role in controlling interface processes such as water repellency. They therefore also play a role in controlling surface and subsurface hydrology as well as the spread of contaminants.

4.3 Sorption (partitioning) processes for organic chemicals

Other important interface processes that are intimately linked to soil architecture (matrix and pore systems) and soil OM quality and quantity in combination with organo-clay complexing are sorption of organic vapors (Amali et al., 1994; Petersen et al., 1994, 1995) as well as dissolved chemicals (de Jonge et al., 2000, 2008). Taking a closer look at gaseous phase sorption in soils can lead to a renewed understanding of soil interface phenomena. Gaseous-phase sorption of non-polar chemicals are like WR highly dependent on soil-moisture conditions, likely making the chemical interface processes in soils highly complicated at the microscale and at present unpredictable at the

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pedon or larger scale. The WR-w curves for the Lundgaard and Ødum soils (marked in Fig. 11) are plotted together with curves relating gaseous phase sorption of the non-polar chemical trichloroethylene with w (Fig. 12). The gaseous phase sorption at zero and very low soil moisture contents takes place directly at the mineral and organic surfaces. While soil-water contents increase, polar water molecules start competing with the non-polar TCE molecules for sorption sites. From oven-dry conditions to around soil-water contents of approximately four-five molecular layers the VOC adsorption capacity decreases non-linearly and drastically (Petersen et al., 1994, 1995) and Henry's Law is not valid. For this non-Henrian region the adsorption/dissolution (Eq. 9) is described by applying a solid/vapor adsorption coefficient, K'_D ($\text{cm}^3 \text{g}^{-1}$), defined from:

$$S_S + S_w = K'_D C \rho_b \quad (9)$$

where S_S is the amount of VOC-vapor adsorbed to soil solids (g cm^{-1}), S_w is VOC-vapor dissolved in water (g cm^{-1}), C is VOC concentration (g VOC cm^{-3}), and ρ_b is the bulk density (g cm^{-3}). In the non-Henrian region between 0 and ~ 4 molecular layers of water coverage, the change in K'_D with changing water content can be predicted by Eq. (10):

$$\log K'_D = (A_0 - \beta)e^{-\alpha w} + \beta \quad (10)$$

where $A_0 = \log K'_D (w=0)$, β is a fitting parameter, and α as a function of β is given by:

$$\alpha = \frac{\ln \left(\frac{A_4 - \beta}{A_0 - \beta} \right)}{w_4} \quad (11)$$

where w_4 is the water content at four molecular layers of water coverage, and $A_4 = \log K'_D (w_4)$. For soil-water contents higher than four molecular layers (Henrian region), the K'_D slightly increases with increasing water contents due to VOC dissolution into soil water. This phenomenon can be described by Eq. (12) assuming the

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applicability of Henry's Law and that water surface sorption and condensation are negligible and correspond to the linear parts of the K'_D - w curves (Fig. 12):

$$K'_D = \frac{K_D}{K_H} + \frac{w}{K_H \varphi \rho} \quad (12)$$

where K_D is the aqueous/solid partition coefficient ($\text{cm}^3 \text{g}^{-1}$), K_H is Henry's constant, φ is the aqueous activity coefficient ($=1$), and ρ is the density of water ($=1 \text{ g cm}^{-3}$).

When moving from the Non-Henry to the Henry region (Fig. 12) – around a molecular water coverage of 4–5, which is also being approximately equal to the wilting point – we seem to reach a crossroad in interface phenomena. The VOC vapour sorption reaches a minimum and around the same point the WR starts increasing. This corroborates with the hypothesis of Or et al. (2007) that partial water coverage of solids surfaces is one of the physical constraints/controls determining numerous interface processes. At the same time the geometry of the bound water (its distribution in films and pore throats) as well as the type of available surfaces (NC, NCC, COC, or NCOC) probably plays an important role regarding interface phenomena (Or et al., 2007; Dexter et al., 2008).

Surface area (total a_s , for example, estimated by EGME measurement or external a_s , for example, estimated by N_2 -BET) appears to be a key player in both vapor sorption and likely also WR, with surface areas of course being closely related to both OM and clay (the finer fractions of the soil matrix). For vapor sorption, clay mainly controls the dry region and OM the wet region. For WR, the interactions between OM and clay together with OM quality, control WR- w curve shape and magnitude. The analysis suggests that both the vapor sorption curve (K'_D - w) and the WR curve (WR- w) may be key interface characteristics towards understanding soil architecture and functions.

The carbon saturation effect is examined further (Fig. 13) with regard to sorption of the dissolved polycyclic aromatic hydrocarbons (PAH) pyrene and phenanthrene. Generally PAHs adsorb strongly to the soil solid phase leaving them unavailable to leaching in the dissolved phase. Sorption coefficients of PAHs are frequently highly

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correlated to the soil OC (see reviews by Pignatello, 1998 and Huang et al., 2003). The OC contents of eight European soils (de Jonge et al., 2008; Celis et al., 2006) are plotted as a function of their clay content (Fig. 14). The soils split into two groups with one above and one below the saturation line as defined by Dexter et al. (2008).
5 The sorption coefficients, K_D , for pyrene and phenanthrene (de Jonge et al. 2008) are significantly correlated to the NCOC fraction of the soils as calculated by Eq. (8), and an improvement on the correlation to soil OC. The sorption coefficients are not correlated to the COC. Again, part of the OC pool seems to be mainly responsible for interface processes within soil inner space.

10 **5 Interrelations of key parameters in soil structure: towards a road map for soil inner space**

5.1 A scientific road map to soil inner space

Soil is the most complex biomaterial on the planet (Young and Crawford, 2004). If we look at soil inner space through a magnifying glass, it seems chaotic. At this scale,
15 the natural intact soil represents an almost unimaginably complex architecture. At first glance, mineral particles, abiotic and biotic organic colloids, “colonies” of particles (aggregates) and biomass (films) appear connected in a random fashion. Constricted by this complicated architecture, water and numerous types of colloids, dissolved chemicals, air, and gaseous compounds move by tortuous routes. By their own translocation
20 and ensuing biophysical processes, the mobile components are constantly changing soil architecture. The first step towards quantifying these not-yet-understood soil phenomena is to envision a scientific roadmap to soil inner space. The map must identify the main bridges and highways for transport and information that are linking key soil constituents and processes and at the same time respecting that any change to a constituent
25 or a process will in itself influence soil architecture and infrastructure.

The pore and particle phases are sources of mobile soil constituents including gases,

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liquids, clay particles, and biotic and abiotic OM (Fig. 14). The mobile constituents travel along tortuous roads and highways formed by complex pore networks, while undergoing translocation processes such as diffusion, convection, dispersion, chemical sorption, and physical straining. External system inputs to soil architecture include
5 management induced energy gradients, climate-induced gradients in moisture matric potential and temperature, and plant-root-induced exudates such as extracellular polymeric substances (EPS) working as biological glues in soil structure formation (Or et al., 2007). The external inputs interacting with the micro-scale biophysical processes cause a variety of not-yet-understood phenomena including creation/breakdown of aggregates,
10 blocking of pore networks by deposited colloids, water repellency and onset of finger-like water flow patterns, inter-connections of previously entrapped air-filled pore space, and onset of gas transport and dispersion. All these processes are intimately linked to the temporal and spatial dynamics of soil inner space architecture, which in turn changes in constant interaction with the biophysical processes.

15 **5.2 Example of a crossroad: Soil-water matric potential and the Dexter clay saturation line**

One interesting crossroad is how a change in soil-matric potential (for example quantified by pF) will affect soil architecture depending on whether the local soil element is above or below the Dexter clay saturation line. If there is a surplus of NCOC and the
20 water potential decreases below 1000 hPa ($pF > 3$), the analyses in this study would imply that water repellency, a more heterogeneous distribution of moisture on the micro-scale, and the occurrence of vapour sorption may be promoted. If instead there is a surplus of NCC, release and redistribution of water dispersible colloids (clay) may occur more easily. Any such threshold-induced change in soil infrastructure will both
25 govern and, in turn, be affected by translocations of biotic and abiotic colloidal particles, water, solutes, air, and gases through phases and across interfaces and thus decide the overall soil functions and human and environmental impact. The first draft of a “Road Map to Soil Inner Space” (Fig. 14) can hopefully guide us towards un-

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derstanding irregular water flow and distribution, preferential gas transport linked to soil architecture, colloid mobilization and chemical-facilitated transport related to soil structure thresholds (healthy sponges versus unhealthy pipes), and moisture and OC thresholds for chemical phase distribution behavior, and finally as the ultimate vision the controls and constraints of self-organization and soil bio(mass)-physical interactions in time and (inner) space.

6 Conclusions

Conclusions that can be drawn on the preliminary results of the Soil-it-is program include:

- 10 – If carefully integrated and interpreted, multi-parameter measurements at scales from aggregates to minimally disturbed bulk soil samples (100 cm³ or larger) may provide valuable information of processes at a lower scale, although we acknowledge the great heterogeneity in soil architecture and processes at the colloid to cluster scale.
- 15 – Novel data analyses of soil-gas transport parameters are presented, including normalization of gas diffusivity data with regard to the WLR model for structureless porous media, and a Brooks-Corey power-law analysis taking into account a percolation threshold for gas diffusivity and introducing the ratio of intra- to interped tortuosity-connectivity factors, m , as a useful indicator for characterizing pore-network architecture.
- 20 – The new analyses supported a hypothesis of possible pipe-like pore network behavior for a soil below the Dexter et al. (2008) clay saturation line while an adjoining, differently managed soil of similar texture but placed above the clay saturation line exhibited a more sponge-like pore network behavior. The pipe-like pore structure of the Low-C soil is seen as a result of dispersion-cementation cycles of clay

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not complexed by organic carbon (NCC), which is supported by high dispersibilities of field moist soil and high mechanical strengths of dry aggregates.

- 5 – The Dexter et al. (2008) clay saturation concept in combination with distribution and intensity of soil water may also in part govern functional soil architecture with regard to hydrological interface processes such as water repellency and fingered flow. For example, the area under the water repellency curve (water repellency versus gravimetric soil-water content) was positively correlated to non-clay-complexed organic carbon (NCOC) for a number of differently textured and managed Danish soils.
- 10 – Also for interface processes governing the fate of environmental impact chemicals such as sorption of volatile organic chemicals and PAHs, both NCOC and soil-moisture status were shown to play a key role in the phase partitioning of both volatile vapors and PAHs.
- 15 – Based on these preliminary results, we envision a road map to soil inner space where important crossroads for key parameters (e.g. NCOC and soil-water matric potential) govern the build-up and resilience or breakdown of soil infrastructure and the thresholds for the ability of soils to self-organize after structure breakdown.
- 20 – The road map may hopefully serve as inspiration for a more focused and multidisciplinary effort combining high-resolution measurement methods from soil sciences, hydroponology and engineering disciplines in soil inner space.

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Table 1. Selected soil characteristics for two arable soils with management-derived differences in OM (modified after Munkholm et al., 2001; Schjøning et al., 2002a; Elmholt et al., 2008). Within each row, figures followed by different letters (a, b) are significantly different ($P=0.05$).

Soil characteristics	Management system	
	High-C-input	Low-C-input
Soil org. C, total (mg g^{-1} soil)	19.7 ^b	14.3 ^a
Soil org. C, hot-water extractable ($\mu\text{g g}^{-1}$ soil)	232 ^b	202 ^a
Non-complexed clay (NCC) (mg g^{-1} soil) ⁴	0	51
Water-dispersible-colloids of wet soil (mg g^{-1} clay)	98 ^a	134 ^b
Water-dispersible-colloids of dry aggregates ¹ (mg g^{-1} clay)	20.6 ^b	18.0 ^a
Tensile strength of dry aggregates ² (kPa)	215 ^a	267 ^b
Wet macro-aggregate stability ³ (mg g^{-1} soil)	637 ^a	873 ^b

¹ Averaged across three aggregate sizes, 0.063–0.25, 0.5–1, 4–8 mm.

² Averaged across four aggregate sizes, 1–2, 2–4, 4–8 and 8–16 mm.

³ >0.25 mm aggregates after Yoder-type wet-sieving for two minutes.

⁴ Calculated according to Dexter et al. (2008), not tested statistically.

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Table 2. Soil and system characteristics for soils with clay minerals interacting with OM in a self-organized manner and for soils below a critical threshold of OM.

Soil and system characteristics	Structure self-organization	
	Active	Collapsed
Org. C inputs (crop rotation, manure, etc)	high	low
Soil org. C quantity and quality	high	low
Non-complexed clay (NCC) ^a	very low or none	>0
Water-dispersible-colloids (clay minerals) of wet soil	low	high
Mechanical strength of dry soil	low	high
Complexity of the soil pore system	high	low

^a As defined by Dexter et al. (2008)

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Table 3. Soil characteristics for Jynde vad, Lundgaard, and Ødum soil.

	Clay	Silt	Sand kg kg ⁻¹	Org. C
Jynde vad	0.048	0.053	0.869	0.011–0.028
Lundgaard	0.048	0.132	0.802	0.011
Ødum	0.137	0.264	0.575	0.014

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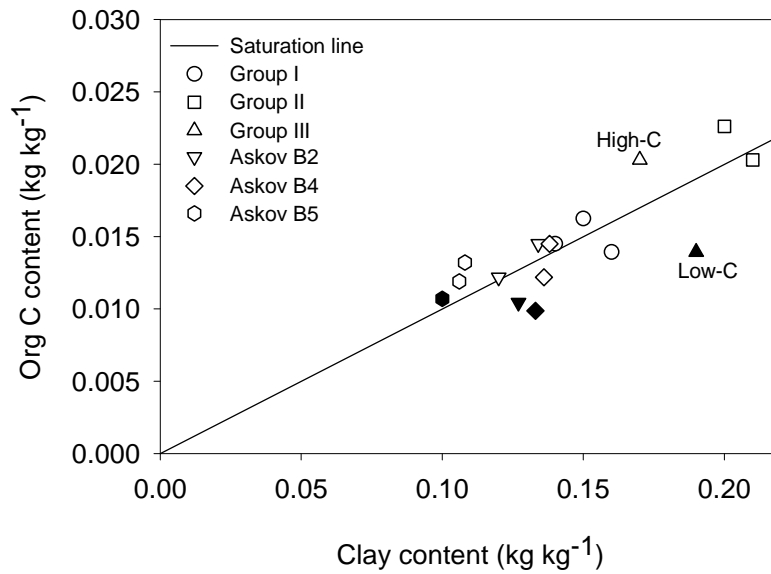


Fig. 1. Relation between the content of clay and OC for a range of Danish soils with different soil management. Results on soil physical characteristics were reported for the Group I through Group III soils by Munkholm et al. (2001) and Schjøning et al. (2002a), the Askov B2 and B4 fields were studied by Munkholm et al. (2002), and the Askov B5 soil by Schjøning et al. (1994). Soils with poor tilth conditions are shown by closed symbols. The soils labelled with “Low-C” and “High-C” are those discussed in detail in Figs. 2 through 6.

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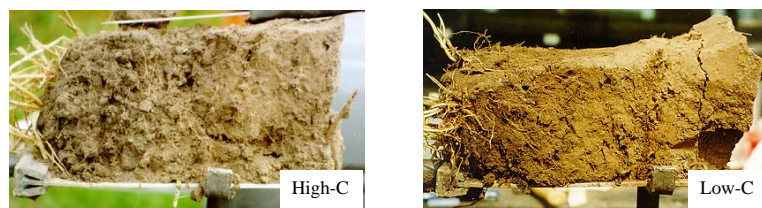


Fig. 2. Photos of the top 30 cm of the High-C soil (left) and the Low-C soil (right) used for evaluation of organic depletion effects on the soil matrix and soil pores (reproduced from Schjøning et al., 2002a).

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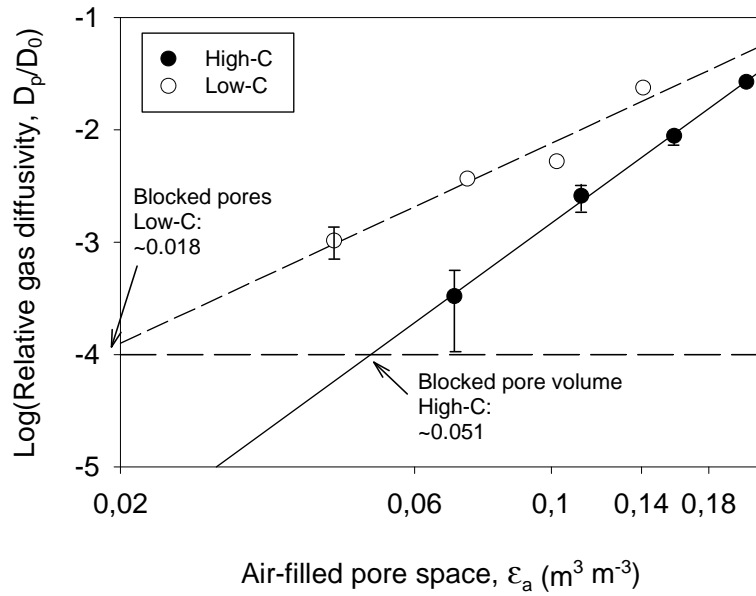


Fig. 3. Gas diffusivity for top layer soil of two arable soils with contrasting contents of management-derived OM (calculated from Schjøønning et al., 2002b). Consult text for details.

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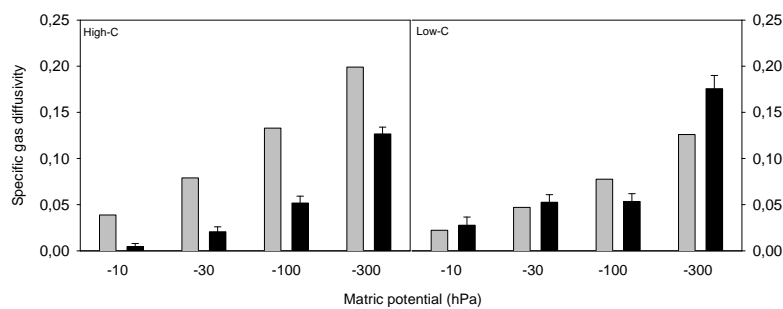


Fig. 4. Specific gas diffusivity ($D_p/(D_0\epsilon_a)$) calculated from measured diffusivities and air-filled porosities (black bars) and from the WLR (Marshall) model with the measured air-filled porosities. Consult text for details. Based on data from Schjøønning et al. (2002b).

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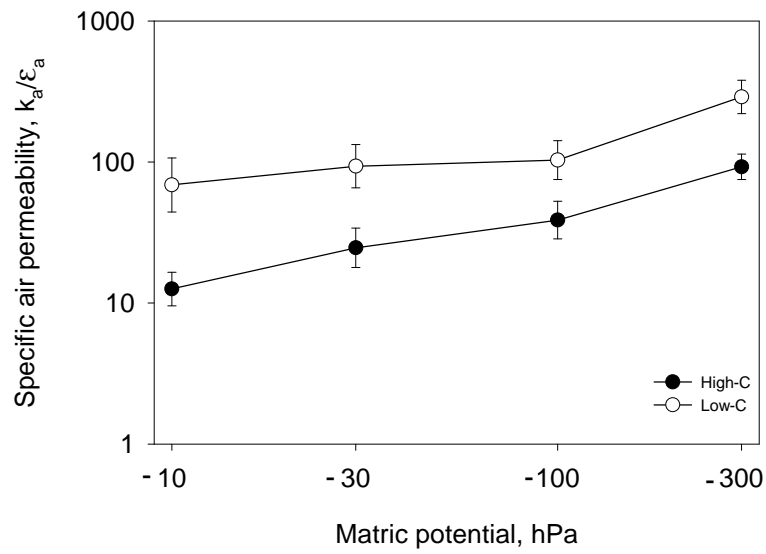


Fig. 5. Specific air permeability (k_a/ϵ_a) at four soil-water matric potentials. Based on data from Schjønning et al. (2002b).

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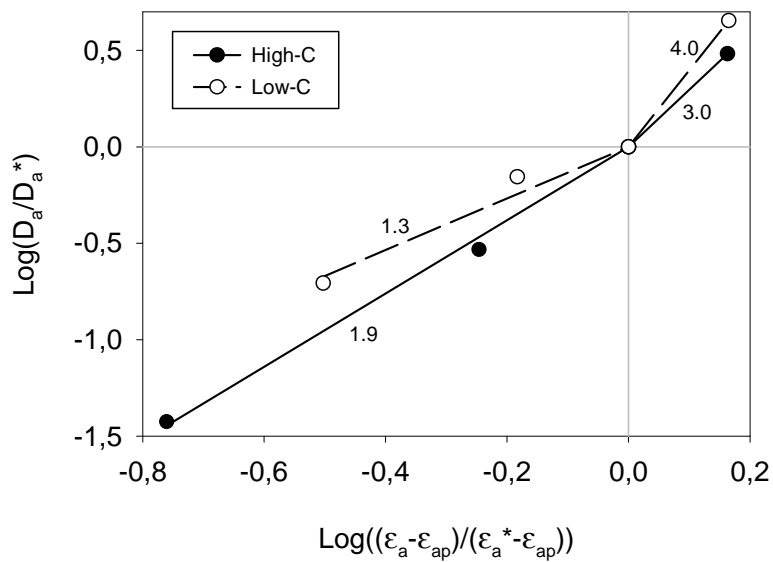


Fig. 6. Normalized relative gas diffusivity, D_p/D_a^* , plotted towards normalized air-filled porosity active in the diffusion process, $(\epsilon_a - \epsilon_{ap})/(\epsilon_a^* - \epsilon_{ap})$, in a log-log presentation as suggested by Moldrup et al. (2001) and with a soil-water matric potential of -100 hPa as the reference point. The figures next to the regression lines give the slopes for the respective parts of the water potential range. Calculated from data of Schjønning et al. (2002b).

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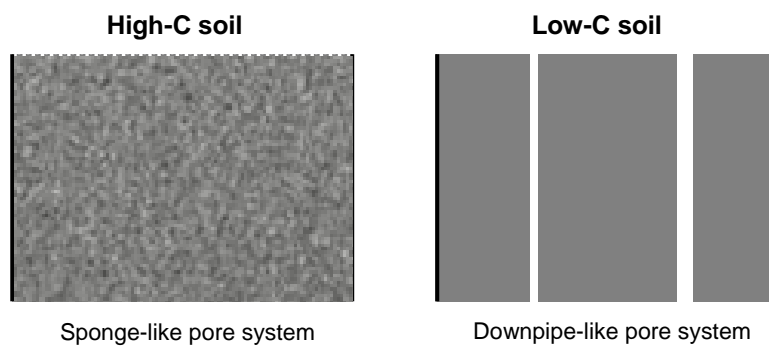


Fig. 7. Conceptual representation of the sponge-like pore system for a high-C soil with active self-organization and a low-C soil, where internal crusting of dispersed clay has created a pipe-like pore system with poor contact between micro- and macropores.

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Fig. 8. Fingered flow phenomena in situ and in laboratory experiments. **(A)** Finger flow in water-repellent sandy soil with permanent grass: 24 h after intensive irrigation the grass layer was removed. Water infiltrates in fingers leaving large sections dry. **(B)** Evidence of finger flow in laboratory hele-shaw irrigation experiments on dry water repellent sandy soil.

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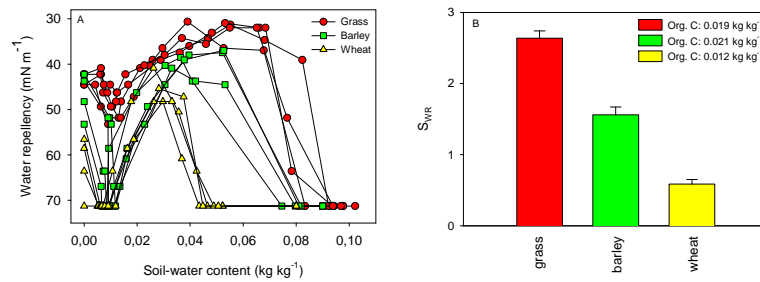


Fig. 9. (A) Soil water repellency versus gravimetric soil-water content curves for three selected crop types grown on the same coarse-textured sandy soil type. Note WR increases with decreasing surface tension (N m⁻¹). Amount and type of soil OM are the two switches influencing the degree of water repellency. Modified from de Jonge et al. (2007). **(B)** Degree of water repellency given as the areas under WR-w curves (Fig. 10a) as a function of Org. C content.

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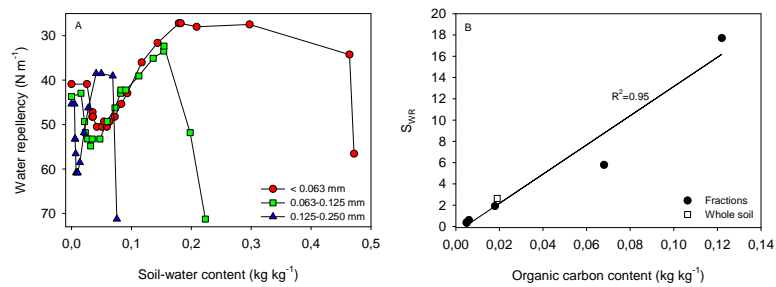


Fig. 10. (A) Soil water repellency as a function of soil-water content for three fractions of a Jynde vad soil. The trapezoidal integrated area under the curves S_{WR} gives the overall degree of water repellency. **(B)** Plotting the trapezoidal integrated area in the WR-w curves, S_{WR} , against OC in the fractions as well as the whole soil shows that the OC content dominates the degree of water repellency.

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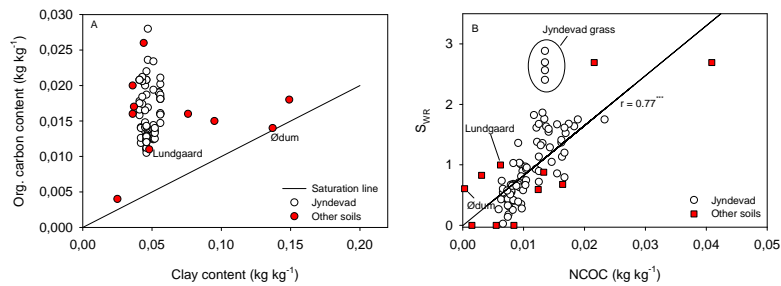


Fig. 11. (A) Relation between the content of clay and OC for numerous treatments of the Jyndeved soil (based on data from de Jonge et al., 2007) as well as 10 additional hydrophilic or hydrophobic soils (based on data from de Jonge et al., 1999). **(B)** The overall WR as determined by the trapezoidal integrated area under the WR-w curves as a function of non-complexed organic carbon (NCOC). The data are for the same soils as in Fig. 11a.

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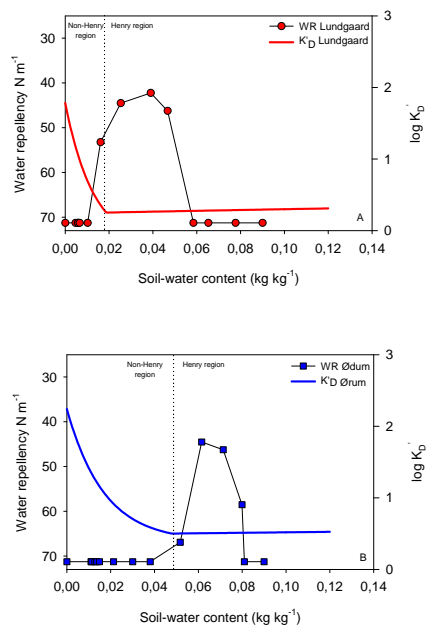


Fig. 12. Curves relating water repellency as well as gaseous phase TCE sorption to soil-water content for **(A)**. Lundgaard (0.048 and 0.011 kg kg⁻¹ clay and Org. C, respectively) and **(B)**. Ødum (0.137 and 0.014 kg kg⁻¹ clay and Org. C, respectively) soils. For gaseous phase sorption the sorption coefficient at low soil-water contents behaves in a non-Henry way (the non-linear part of the curve), where gas molecules adsorb directly to mineral or organic surfaces or even bound water molecules. When there is sufficient water bound to the soil, the sorption coefficients can be predicted from Henry's Law and are dominated by partitioning from the water phase to sorption sites (the linear part of the curves).

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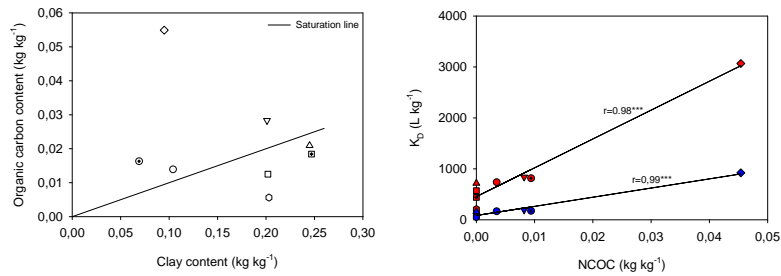


Fig. 13. (A) Relation between the content of clay and OC for eight European soils (modified after de Jonge et al., 2008). (B) The sorption coefficient of pyrene (red) and phenanthrene (blue) as a function of NCOC content in the soils.

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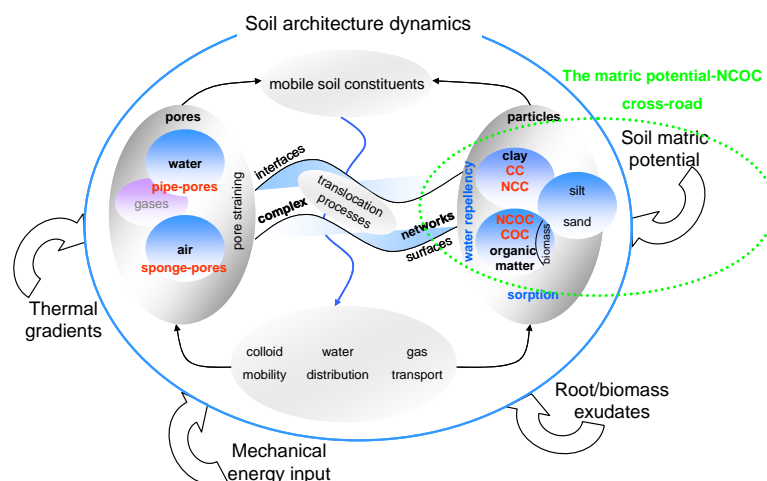


Fig. 14. Schematic road map to soil inner space and some not-yet-understood processes and phenomena. CC: Complexed clay, NCC: Non-complexed clay, COC: Complexed organic carbon, NCOC: Non-complexed organic carbon.

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