

Aus dem Institut für Pflanzenernährung und Bodenkunde
der Christian-Albrechts-Universität zu Kiel

**Optimization of the soil structure in arable crop production
by means of lime (CaCO₃) application**

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*„Und der Zweifel ist tiefste Menschlichkeit, ist der Antrieb zu denken, zu hinterfragen,
zu suchen“ Siddhartha, Hermann Hesse, 1922*

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LIST OF ABBREVIATIONS AND SYMBOLS

ABBREVIATIONS

AAS	atomic adsorption spectrophotometer
AC	air capacity [vol.%]
ANC	acid neutralization capacity [mol H ⁺]
BNC	base neutralization capacity [mol H ⁺]
C	carbon [%]
CCE	calcium carbonate equivalence [%]
CEC	cation exchange capacity [cmol _c kg ⁻¹]
CEC _{pot} , CEC _{eff}	potential CEC, effective CEC [cmol _c kg ⁻¹]
DLVO	Derjaguin-Landau-Verwey-Overbeek
ext.	exterior
FP	fine pores
int.	interior
N	nitrogen [%]
nPAW	not plant available water [vol.%]
PAW	plant available water [vol.%]
pH	pondus hydrogenii, negative of base 10 logarithm of concentration of H ⁺ ions [-]
SOC	soil organic carbon
SOM	soil organic matter [g kg ⁻¹]
TP	total pore volume [vol.%]

LIST OF ABBREVIATIONS AND SYMBOLS

SYMBOLS

c	cohesion [kPa]
C_2	pore continuity index
E_s	erosive strength
ε_a	air-filled porosity [$\text{cm}^3\text{cm}^{-3}$]
k_a	air conductivity [cm d^{-1}]
k_{ap}	air permeability [m^2]
k_s	saturated hydraulic conductivity [cm d^{-1}]
k_u	unsaturated hydraulic conductivity [cm d^{-1}]
ξ	zeta potential [mV]
ρ	aggregate density [g cm^{-3}]
ρ_B	bulk density [g cm^{-3}]
σ_n	normal stress [kPa]
σ_{pc}	pre-compression stress [kPa]
τ	shear strength [kPa]
ϕ	angle of internal friction [$^\circ$]
χ	parameter between 0-1 related to the degree of saturation of the soil
Ψ_m	matric potential [kPa]

1 Summary

Mechanical interventions in the form of soil cultivation on agriculturally used land lead to changes in mechanical, hydraulic and biological soil properties that affect the soil structure. As a result of the applied stress, the pore functions are impaired, and the water and air balance are negatively affected. In order to establish a more stable and at the same time looser soil structure, an optimal supply of lime to the soils is essential. Important chemical (pH value, nutrient sorption (CEC)), biological (microbial activity) and physical (water and gas transport) properties on soils of different textures can be changed in such a way that this leads to an increase in nutrient storage and availability for plants and makes an important contribution to soil protection. The present study therefore deals with the effect of different levels of lime (CaCO_3) on various soil properties. Seven (six of them in this thesis) field trials were established with three different rates of lime in various soil regions in Germany, with a wide range of different textures, clay contents and soil organic matter. Disturbed and undisturbed soil material was taken from the topsoil (0-30 cm) during the annual sampling (2017-2020) to quantify the stability and structural changes of the soil on the mesoscale (precompression stress and frame shear tests as well as crushing tests). Furthermore, the bulk density, pore size distribution, air- and saturated water conductivity were measured, and the chemical properties of the equilibrium soil solution were determined. The stability but also the structural change of a soil depends on the complex interaction and a wide range of soil properties and mechanisms. Depending on the clay and organic matter content, the soils show different reactions to lime additions and thus to the release of Ca^{2+} and carbonates regarding particle interaction. In the stability measurements on the mesoscale, there are already overlaps of a possible lime effect by external factors, such as the annual tillage, which complicates transferability of the Ca^{2+} effect. Factors such as aggregation and time of sampling as well as associated swelling and shrinkage processes also influence the effect or accessibility of Ca^{2+} to the exchange places. Nevertheless, a (time-dependent) correlation between changes in the pore system and stability as well as structure-changing processes can be observed during frame shear tests. While an increased addition of lime to sandy soils can lead to both a (short-term) improvement in water retention and saturated water conductivity and shear strength, such an effect can be

seen with a delay for soils containing considerable amounts of clay. The rearrangement, interlinkage and connection of soil particles improve the mechanical stability of soil aggregates, shear resistance and the stability of the pore system. Liming reduces the swelling capacity of clay-rich soils and can thus mitigate structural weaknesses of these soils. Therefore, the trafficability of such soils can be improved by liming.

2 Zusammenfassung

Mechanische Eingriffe in Form der Bodenbearbeitung auf landwirtschaftlich genutzten Flächen führen zu Veränderungen von mechanischen, hydraulischen und biologischen Bodeneigenschaften, die sich auf die Bodenstruktur auswirken. Als Folge der Belastung kommt es zu Beeinträchtigungen der Porenfunktionen und damit zu einem negativ beeinflussten Wasser- und Lufthaushalt. Um eine stabilere und gleichzeitig lockere Bodenstruktur zu etablieren, ist eine optimale Versorgung der Böden mit Kalk unerlässlich, es können wesentliche chemische (pH-Wert, Nährstoffsorption (KAK)), biologische (mikrobielle Aktivität) und physikalische (Wasser- und Gastransport) Eigenschaften auf Böden unterschiedlicher Struktur so verändert werden, dass dies zu einer Steigerung der Nährstoffspeicherung und –Verfügbarkeit für Pflanzen führt und einen wichtigen Beitrag zum Bodenschutz liefert. Die vorliegende Arbeit befasst sich daher mit der Wirkung unterschiedlich hoher Kalkgaben (Kohlensauer Kalk, CaCO_3) auf verschiedene Bodeneigenschaften. Es wurden sieben (davon sechs in dieser Thesis) Kalksteigerungsversuche in unterschiedlichen Bodenregionen Deutschlands angelegt und beprobt, wobei eine breite Spanne unterschiedlicher Texturen, Tongehalten und Humusgehalten vorliegt. Es wurde bei den jährlichen Probenahmen (2017-2020) gestörtes und ungestörtes Material aus dem Oberboden (10-30 cm) entnommen, um die Stabilität bzw. Strukturveränderungen des Bodens auf der Mesoskala (Drucksetzungs- und Kastenscherversuche, Crushing-Test) zu quantifizieren. Weiterhin wurden die Lagerungsdichte, Porengrößenverteilung, Luft- und gesättigte Wasserleitfähigkeit gemessen und die chemischen Eigenschaften der Gleichgewichtsbodenlösung erfasst. Die Stabilität aber auch die Strukturveränderung eines Bodens hängt vom komplexen Zusammenspiel und einer Vielzahl von Bodeneigenschaften und Mechanismen ab. Je nach Ton- und Humusgehalt zeigen die

Böden hinsichtlich der Partikelinteraktion unterschiedliche Reaktionen auf die Kalkgaben und somit auf die Freisetzung von Ca^{2+} und Karbonaten. Bei den Stabilitätsmessungen auf der Mesoskala kommt es bereits zu Überlagerungen eines eventuellen Kalkeffektes durch externe Faktoren, wie zum Beispiel die jährliche Bodenbearbeitung, sodass die Übertragbarkeit der Ca^{2+} -Wirkung teilweise erschwert wird. Auch Faktoren wie Aggregation und Zeitpunkt der Probenahme sowie damit verbundene Quellung- und Schrumpfungsprozesse beeinflussen die Wirkung bzw. Zugänglichkeit von Ca^{2+} an den Austauscher. Dennoch kann mitunter ein (zeitlich abhängiger) Zusammenhang zwischen Veränderungen des Porensystems und der Stabilität sowie strukturverändernde Prozesse im Verlauf von Scherversuchen festgestellt werden. Während eine gesteigerte Kalkgabe bei sandigen Böden sowohl zu einer (kurzfristigen) Verbesserung der Wasserretention und der gesättigten Wasserleitfähigkeit und der Schergeraden führen kann, ist eine solche Wirkung bei tonigerem Material verzögert zu erkennen. Die Umlagerung, Vernetzung und Verbindung von Bodenteilchen verbessert die mechanische Stabilität von Bodenaggregaten, die Scherfestigkeit und die Stabilität des Porensystems. Die Kalkung reduziert die Quellfähigkeit von tonreichen Böden und kann so strukturelle Belastungen dieser Böden besser abmildern. Daher kann die Befahrbarkeit der untersuchten Böden durch Kalkung verbessert werden.

3 Introduction

Agroecosystems are modified worldwide to enhance their capacity for food production, while they must be sustained economically, environmentally and socially (Warkentin, 2001). In the light of climate change associated with land degradation (Olsson et al., 2020) and a growing world population (FAO et al., 2019), the fate of soils in agroecosystems has recently come more into focus during the latest years. Land degradation processes are caused by human activities due to unsustainable management. Consequences such as wind and water erosion, destruction of soil structure and loss of soil organic matter, which threaten soils functions and thereby lead to economic losses (Olsson et al., 2020).

3.1 Soil structure and aggregation

Soil structure is defined by the spatial arrangement of particles and pores (Díaz-Zorita et al., 2002; Oades, 1984b), and its formation and development results from the rearrangement of particles due to capillary forces as well as from the flocculation and cementation of mineral particles with inorganic and organic substances (Oades, 1984b; Six et al., 2000; Tisdall and Oades, 1982). Climate strongly influences the soil type and the degree of soil aggregation, because of changes in soil matric potential, soil temperature, biological activity and chemical composition of the soil solution. Drying and wetting cycles change the intensity of bonding between particles, because drying of soils decreases the pore water pressure, whereas water menisci pull mineral particles together, resulting in an increased number of contact points, and thus, increased soil cohesion (Horn et al., 1994; Horn and Smucker, 2005; Peng et al., 2007). In addition to the shrinking and swelling processes, Edwards and Bremner (1967) highlighted the consequences of interactions of polyvalent cations and organic ligands with mineral surfaces which initiate the formation of organo-mineral microaggregates (20-250 μm). Tisdall and Oades (1982) defined the interaction of aggregates and soil organic matter with a concept of aggregate hierarchy. This involves different binding partners acting at different hierarchical levels of the aggregation. Free primary particles and silt-sized aggregates (< 2-63 μm) are transformed by binding agents to form stable microaggregates (20-250 μm) (e.g., humified organic matter, roots exudates, polyvalent cation complexes (e.g. Ca^{2+})).

(Bronick and Lal, 2005), oxides (Six et al., 2000) and highly disordered aluminosilicates (Tisdall and Oades, 1982)). These stable microaggregates are in turn bound together into macroaggregates (> 250 μm) by e.g., fungal hyphae and fine roots (Fig. 3-1). Oades (1984b) proposed a modification to the aggregate hierarchy concept of Tisdall and Oades (1982) by theorizing the formation of microaggregates within macroaggregates as a result of several processes such as the reorientation of clay particles around deposited organic debris (Jastrow, 1996). The strength of organo-mineral aggregates depends on the surface area of the involved mineral particles (Kaiser and Guggenberger, 2003). The interactions between roots and mineral particles which also affect aggregation via the release of root exudates such as mucilage (Czarnes et al., 2000; Tisdall and Oades, 1982) and even bacteria provides binding agents for the formation of microaggregates (Feeney et al., 2006). The variation in soil texture influences the aggregation process (Dörner et al., 2010) because in coarse sandy soils hyphae are able to cross-link the existing sand particles to form stable aggregates. In clayey and loamy soils, soil aggregation is affected by bacteria and fungi as well as their products (Six et al., 2004). Further, the aggregates in fine-textured soils can be subject to swelling and shrinking processes (Peng et al., 2007), modified by compression and compaction, or broken down by plowing, excavations or other anthropogenic processes on the field.

The stability, size distribution and spatial organization of soil aggregates influence the pore geometry (Lipiec et al., 2009), and the overall mechanical stability of the soil (Kay, 1990). Soil aggregation preserves pore continuity, pore connectivity, tortuosity, gas and water transport, and aggregates may function as storage medium for water, nutrients and organic matter constituents or habitat for microorganisms (Baumgarten and Horn, 2013).

Aggregates persist as long as the soil strength is higher than the given load or shrinkage forces, which also includes the effect of chemical and biological consecutive stabilizing (Horn and Peth, 2009). Soil strength increases with aggregation: coherent < prismatic < blocky < crumbly < subangular (Hartge et al., 2014; Hillel, 2004).

The opposite of aggregation is soil fragmentation, which refers to the process of breaking soil apart into incomplete fragments (Díaz-Zorita et al., 2002). Arshad et al. (1997) described the transition of the soil into a transient unstable mass called clods

after the disturbance due to tillage. Anthropogenic activities largely influence the soil quality and can potentially result in reduced soil fertility, water infiltration and nutrient buffering functions (Beylich et al., 2010; Horn et al., 1995; Keller et al., 2013). The disruption of aggregates during tillage causes enhanced mineralization of previously protected soil organic carbon (SOC) followed by increasing emissions of greenhouse gases such as carbon dioxide (CO₂) (Caires et al., 2006; Sheehy et al., 2015; Six et al., 2000) (Fig. 3-1). The protection of SOC by entrapment in aggregates and the aggregate formation can be enhanced by an increase of microbial activity and root growth, which results in higher proportions of organic compounds and polysaccharides in the soil (Briedis et al., 2012a). Physical protection and storage of carbon or nutrients in aggregates can reduce greenhouse gas emissions (Mordhorst, 2013), act as a sink and thus be an important factor to mitigate climate change.

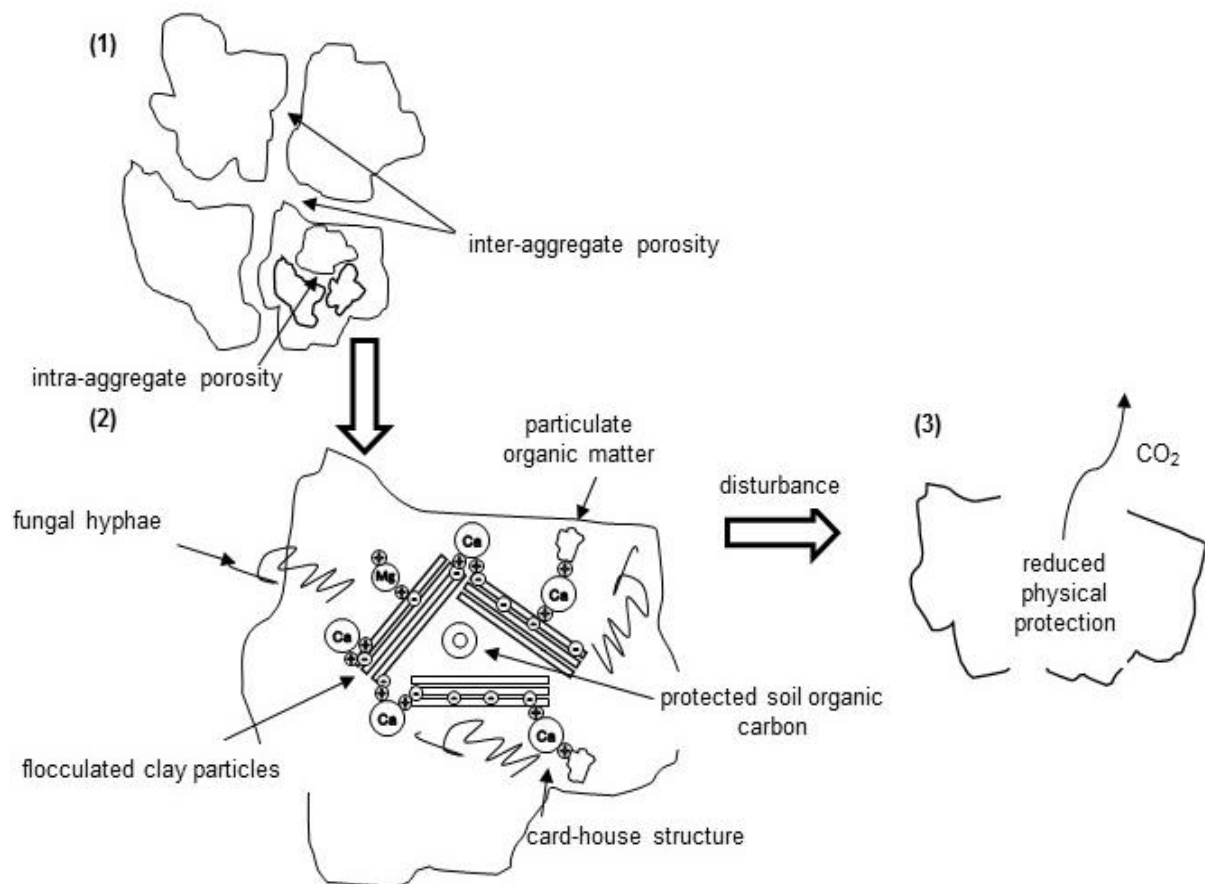


Fig. 3-1: Macro-aggregate and (2) Micro-aggregate with flocculated clay particles. (3) Disturbance of an aggregate (own figure).

When a soil is mechanically loaded, forces referred to as pressure or tension in relation to a defined area act on the soil and cause stresses inside the soil. The effective stress occurs between the particles in the soil (Mitchell and Soga, 2005b). In the three-phase system of the soil, the effective stress equation must consequently include the solid, liquid and gaseous phases, all considered with respect to the individual stress components according to Bishop (1960). In this case, a χ -factor, describing the surface area of the pore water or, conversely, the surface area of the air-filled pores at an imaginary cross-section through the pore system of the unsaturated soil, must be included in the effective stress equation (Likos, 2014) (eq. 3–1).

$$\sigma' = \sigma - \chi u_w \quad \text{eq. 3-1}$$

with

σ' = effective stress (kPa)

σ = total stress (kPa)

χ = degree of water saturation; ($\chi = 1$) and complete dryness ($\chi = 0$)

u_w = pore water pressure (kPa)

Remolded and reworked soils are more affected by loading and shear forces than those with an intact structure (Horn et al., 1994). The modifications of the soil pore structure during loading depend on several factors, including strength of pre-existing structure units as well as amount and type of compaction or remolding. In general, two processes of deformation can occur in the soil: elastic deformation (the internal soil rigidity is not exceeded, the deformation is reversible), and plastic deformation (Horn and Fleige, 2003). During loading, the soil structure can carry effective stress but, if the load applied to the soil exceeds the internal soil strength, the structure is disrupted, effective stresses are reduced, and the internal soil strength decreases.

Soil shear strength, as defined by the resistance of soil towards shear deformation under mechanical impact, can be assessed from the shear parameters angle of internal friction and cohesion. The shear strength defines the ability of the soil structure to withstand shear forces at a given stress applied (Horn, 1990). The alterations of soil structural stability due to mechanical impacts and compaction as well as the naturally occurring fluctuations (shrinking and swelling cycle affected by changes in the matrix potential) during the course of a year are well documented (Hartge and Horn, 2009; Horn, 2004; Horn and Lebert, 1994). Compaction occurs when the soil's inherent

strength is exceeded by agricultural machinery traffic especially on wet and/or weak soils and increasing traffic intensity (heavier machinery and/or higher traffic frequency) increases the susceptibility to irreversible soil deformation (Duttmann et al., 2014; Horn and Peth, 2012; Soane and van Ouwerkerk, 1994). Vertical pores, such as root and earthworm channels or primary shrinkage cracks are less sensitive to compression than oblique or horizontal pores (Schäffer et al., 2008). Nevertheless, if wheeling is repeated over years with stresses exceeding soil strength, the aggregates formed and stabilized by chemical and biological binding agents are not able to withstand this exposed stress, and a horizontal rearrangement of pores between plates including an increased horizontal anisotropy of pore functions will occur (Riggert et al., 2017). Aggregate disruption, homogenization and kneading of the soil, and a coinciding decline of the angle of internal friction as well as cohesion are the consequences. These processes also lead to the complete blockage of formerly connected transport paths and thereby inhibit water and gas fluxes (Horn, 2003; Huang et al., 2021). Increasing bulk density also impedes plant root growth which, along with reduced aeration, limits the suitability of the soil for crop production. Soil characteristics such as hydraulic conductivity, plant available water capacity as well as air capacity are negatively affected by loading and shearing, e.g. induced by traffic on arable soils, because each rearrangement of soil particles results in an alteration of pore spaces in the soil (Gupta et al., 1989). Furthermore, soil deformation alters the spatial pore arrangement (Kuncoro et al., 2014; Neugebauer, 2015) and the pore size distribution with a reduction of wide and narrow coarse pores (Gebhardt et al., 2009). The resulting reduced pore continuity has negative effects on e.g. soil aeration (Dörner, 2005; Reszkowska et al., 2011). Consequences of soil degradation are erosion, surface runoff, with an associated loss of nutrients especially in arable land (Mitchell and Soga, 2005a).

3.2 Process of soil acidification

Soil acidification is a slow, continuous natural process that occurs during pedogenesis. Acidic soils are common in areas where soils have developed over long, geologic periods and under climatic conditions where precipitation exceeds evapotranspiration (Rengel, 2011; Riedel and Weber, 2020). When protons, H^+ , are introduced by atmospheric inputs or released by internal substance transformations in the soil, they

react with the buffer substances available in the soil (Vogel et al., 2020). Acid inputs from precipitation include the strong acids H_2SO_4 and HNO_3 , originating mainly from anthropogenic emissions (Bolan et al., 2015). The release of H^+ ions in the soil occurs (1) from H_2CO_3 formed as a result of the reaction of H_2O with CO_2 released from aerobic respiration of soil organisms, (2) in the course of nutrient uptake by plants, wherein H^+ ions are released in exchange for dissolved nutrient cations (Ca^{2+} , Mg^{2+} , K^+ , NH_4^+), (3) from the release of organic acids by plants and soil microorganisms for nutrient mobilization, (4) from the oxidation of NH_4 (released by microbial N-mineralization or applied with fertilizer) to NO_3 (Goulding, 2016; Kunhikrishnan et al., 2016), (5) from the oxidation of reduced sulfur, manganese and iron compounds.

The more CO_2 is formed in the soil gaseous phase, the more H_2CO_3 arises and the more H^+ ions are released to the soil solution. In addition to nutrient uptake by plants, leaching of nutrient cations (Ca^{2+} , Mg^{2+} , K^+ and Na^+ ions) leads to additional acidification (Haynes and Swift, 1986). The amount of leaching losses depends on the soil type (light soils more than heavy soils), on the type and duration of planting (vegetation cover prevents leaching), as well as on the amount and intensity of precipitation (seepage water amount). The pH-buffering capacity of mineral soil is mainly based on carbonates, clay minerals, Mn, Al and Fe oxides or hydroxides and organic matter (Goulding, 2016). In carbonate-rich soils, dissolution of carbonates with consumption of protons is the dominant buffering system (pH between 8.6 and 6.2). Of the greatest ecological importance is silicate buffering – the weathering of primary silicates, which takes place under all pH conditions and which is the dominant buffer reaction in soils free of carbonates at $\text{pH} > 5$ (Ulrich and Sumner, 1991) resulting in the release of nutrients (Ca, Mg, K, Fe, Mn) into the soil. Clay minerals with permanent negative charge and high base saturation can act as fast pH buffers in the rhizosphere, among other places, by cation exchange (between pH 5.0 and 4.2). In this process, free protons are adsorbed on the surfaces, thereby buffering the pH of the soil solution. Solids with variable surface charge (e.g. humic substances, edges of clay minerals, surfaces of oxides and hydroxides) also absorb protons in the case of acid inputs and thus act as pH buffers (Cherian and Arnepalli, 2015). At pH values below 4.2, Al-hydroxy-groups of clay minerals are protonated, and part of the Al^{3+} released during weathering of aluminosilicates is exchangeably adsorbed at cation exchange sites,

which become increasingly saturated with Al^{3+} (in $[\text{Al}(\text{H}_2\text{O})_6]^{3+}$). The exchangeable Al^{3+} is in equilibrium with dissolved Al^{3+} in the soil solution (Wellbrock et al., 2016; Zehetner, 2004). High Al^{3+} concentrations in the soil solution can be toxic for roots and prevent nutrient uptake (Ulrich and Sumner, 1991). The better the soil is buffered (within a certain buffer range), the slower the pH will decrease with the same addition of H^+ ions. In addition to other factors such as aggregation and thus accessibility of surfaces, the decrease of the pH depends on the reaction kinetics and the amount of buffer substances present that act in certain buffer ranges.

The parameter acid neutralization capacity (ANC) represents a capacity parameter of soil alkalization, while base neutralization capacity (BNC) represents a capacity parameter of soil acidification (Ulrich and Sumner, 1991; van Breemen et al., 1983). ANC and BNC depend on the amount of buffer substances in the soil as well as clay content and amount of soil organic matter (Vogel et al., 2020). They represent a quantitative measure of the buffering capacity of soils against bases and acids, respectively. ANC is the amount of acids (e.g., in mol H^+) needed to lower the pH of a (soil) system to a reference pH, while BNC represents the amount of bases needed to raise the pH of a (soil) system to a reference pH. Thus, with increasing acidification, ANC decreases and BNC increases. In agricultural systems, the BNC determines, for example, the amount of lime application necessary to effectively raise the pH (lime requirement) (Blume et al., 2016a; Vogel et al., 2020). The increase in pH after liming of acidic soils leads to an increase of the negative variable charge and thereby an increase in the CEC (cation exchange capacity). The CEC_{eff} (effective cation exchange capacity) refers to the sum of the permanent and variable negative charges on the sorption complex of a soil and is determined from the sum of charge equivalents represented by the main exchangeable cations (Ca^{2+} , Mg^{2+} , Na^+ , K^+ , Al^{3+} , H^+) at the current pH of the soil. The cations Ca^{2+} , Mg^{2+} , Na^+ and K^+ are often referred to as base cations because the hydroxides of these cations (e.g. NaOH) are strong bases. The cations Al^{3+} and H^+ are called acid cations because H^+ is exchangeable in the soil solution and Al^{3+} is formed with water molecules in a hexaquoocomplex ($[\text{Al}(\text{H}_2\text{O})_6]^{3+}$) and releases protons in aqueous solutions through hydrolysis (Blume et al., 2016; Zehetner, 2004). The CEC depends on clay content, type of clay minerals, humified organic matter content and soil pH (Rengel, 2011). Vermiculite and smectite, despite

lower charge density, have a higher CEC than illite. In these clay minerals, both the outer surfaces and the interlayers are accessible to cation exchange. In illites, however, the CEC is lower because most of the permanent negative charges in the interlayers are balanced by non-exchangeable K^+ ions (Lambooy, 2013). The clay minerals kaolinite and halloysite have an extremely low permanent negative charge and the exchange of cations takes place through variable charge at the edges and basal surfaces of the clay minerals (van Alfen, 2014).

Liming is used to influence the acid-base state of the soil. This is of great importance as soil acidity affects the availability of nutrients in the soil and for plants and the cation exchange capacity in the soil.

3.3 Lime application in Germany

Liming is a standard measure on farming lands to achieve the pH that is considered optimal for the given organic matter content, soil texture and type of use (arable land or grassland) (VDLUFA, 2000a). For mineral soils, the percentage of sand reduces both the lime requirement and the initial soil pH at which lime is required. Soils with a higher clay and organic matter content have the greatest lime requirement to compensate for soil acidity (Goulding, 2016), because with increasing amount of clay and organic matter, the soil has a high BNC (Vogel et al., 2020), and thus, a high proportion of bonded acidity, i.e. exchangeable protons at the exchange sites in the soil. At the same time, the increased pH due to lime application results in an increased CEC, and thus, a higher sorption capacity for nutrients in the soil.

The calcium carbonate equivalence (CCE) is the neutralizing value of a liming material compared to pure calcium carbonate ($CaCO_3$). The amount of liming material needed to correct soil acidity depends on the neutralization value of the liming material and the pH buffering capacity of the soil (VDLUFA, 2000a). The range of liming materials that are used in agriculture vary in their ability to neutralize the acidity. These include calcitic limestone ($CaCO_3$) (CCE = 100 %), dolomitic limestone ($CaMg(CO_3)_2$) (CCE = 109 %), slaked lime ($Ca(OH)_2$) (CCE = 136 %), quicklime (CaO) (CCE = 179 %) and slag lime ($CaSiO_3$) (CCE= 60–90 %). The liming materials (oxide, carbonate, silicate) differ in their solubility and act differently, which depends on the pH value (buffer range) in the soil. Lime forms containing silicates ($CaSiO_3$) and

carbonates (CaCO_3) are acid-soluble, i.e. the lower the pH value, the better the solubility (Lalande et al., 2009). Nevertheless, CaCO_3 is considered an important buffer substance in the soil (Clough and Skjemstad, 2000). Although the applied lime does not dissolve immediately in water, it acts over a longer period by buffering acid inputs and gradually releasing structurally effective calcium. At pH values > 6.2 , only water-soluble oxides such as CaO or mixed limes like $\text{Ca}(\text{OH})_2$ with hydroxides are readily available (Schachtschabel and Hartge, 1958).

VDLUFA (2000a) defines five lime supply classes for mineral soils. Classes "A" and "B" represent soils with a poor soil structure, reduced nutrient availability, mobilization and increased plant availability of heavy metals, and consequently serious yield losses. In these acidified soils, lime application is the most important fertilization measure to recover the productivity (= recovery liming), but also to protect the soil against erosion, crusting and compaction (Jacobs et al., 2018) and to achieve the optimal lime supply level "C". However, even where soils presently are at an optimal supply class (C) with a good soil structure and availability of nutrients, it may still be important to support them with regular lime applications to maintain the optimal pH-range (= maintenance liming). In the lime supply classes "D" and "E" an application with lime is not necessary, because an increased pH can lead to a fixation of trace elements and also to reduced plant yields. Soils of these classes occur where calcium is present in the parent rock, for example, in loess-, lime- and Keuper-weathered soils. Within a year, about 400-600 kg/ha CaO-equivalents are either absorbed by roots or leached out by infiltration with water (Rechsteiner and Keller, 2019). Therefore, lime application is required on arable soils at least every three years, to maintain the optimal nutrition status and pH value as recommended by VDLUFA (2000a). The calculation is explained for one of the study sites analyzed in this thesis:

Neubrandenburg, silty sand, clay content: 11 g kg^{-1} , and organic carbon content: 7 g kg^{-1} (0-30 cm)

- Initial pH: 5.8
- Soil group: 3
- Recommended pH: 6.1-6.7
- Lime supply according to VDLUFA (2000b): $25 \text{ CaO dt ha}^{-1}$
- Applied CaCO_3 : 48 % CaO

- Recommended lime supply according to VDLUFA (2000a):
 $25 \times 100 / 48 = 52.1 \text{ dt ha}^{-1}$

In recent years, the regional agricultural authorities have warned about the problems associated with acidification and pointed out the importance to neutralize acid soils in the arable lands of Germany, also for food security (Lorenz et al., 2020). For Thuringian soils, 20 % of deficiency symptoms in plant nutrients are due to an insufficient application of lime (Kerschberger, 2008). Soils that have a low organic matter content and have developed from degraded shale weathering, lower and middle red sandstone, gneiss and porphyry as well as diluvial and alluvial soils often have a higher sand percentage, and thus, a higher risk of acidification because of a low acid neutralization capacity. Lausen (2012) stated that lime application is often neglected in agricultural land use. Currently, 58 % of the arable sandy soils in Schleswig-Holstein are undersupplied with lime. Clay and silt soils have a higher demand for lime supply due to their high sorption and buffering capacity, but even these soils are about 50 % undersupplied in Schleswig-Holstein. In Saxony-Anhalt, more than 25 % of arable soils are undersupplied and have an urgent need of lime (Stolpe and Schimpf, 2017). A statistical evaluation of the lime supply of Bavarian soils has also indicated the need for increased lime fertilization. Here, soil samples were taken between 2014 and 2019 and 28 % of these tested soil samples had to be classified in a "low" supply class (Reisenweber, 2020). A pH-survey in Germany by Jacobs et al. (2018) showed that only 35 % of the arable soils and 24 % of the grassland soils were in the optimum range, whereas the pH of approximately 42 % of the mineral soils under arable farming and 57 % of the grassland soils was too low.

3.4 Effects of lime application in soils

Under humid climatic conditions, soil acidification occurs gradually due to continuous input or formation of acids but is also largely influenced by the acidification of the rhizosphere by plant roots to take up nutrients (Bolan et al., 2015). The pH value of the soil is subject to fluctuations over time and can exhibit considerable spatial variability (Zehetner, 2004). Thus, lime application on arable land is a worldwide practice to overcome the problems associated with acidic and/or structurally damaged soils (Bolan et al., 2015). Liming has several benefits including enhancement of soil

aggregate stability and surface charge (an increasing pH increases the net negative charge (Haynes and Naidu, 1998)), mobilization of nutrients (molybdenum and phosphate (Ruhberg, 2017)) and immobilization of toxic elements (Kunhikrishnan et al., 2016)). To be effective, a liming material must displace hydrogen, aluminum, and manganese ions from exchange sites of soil colloids, neutralize H^+ and precipitate Al^{3+} and Mn^{2+} in soil solution (Helyar and Porter, 1989). The dissolution of lime ($CaCO_3$) applied to soil consumes H^+ ions from the soil solution (increasing of the pH) and releases Ca^{2+} -ions and HCO_3^- (Wisotzky, 2011) (Fig. 3-2).

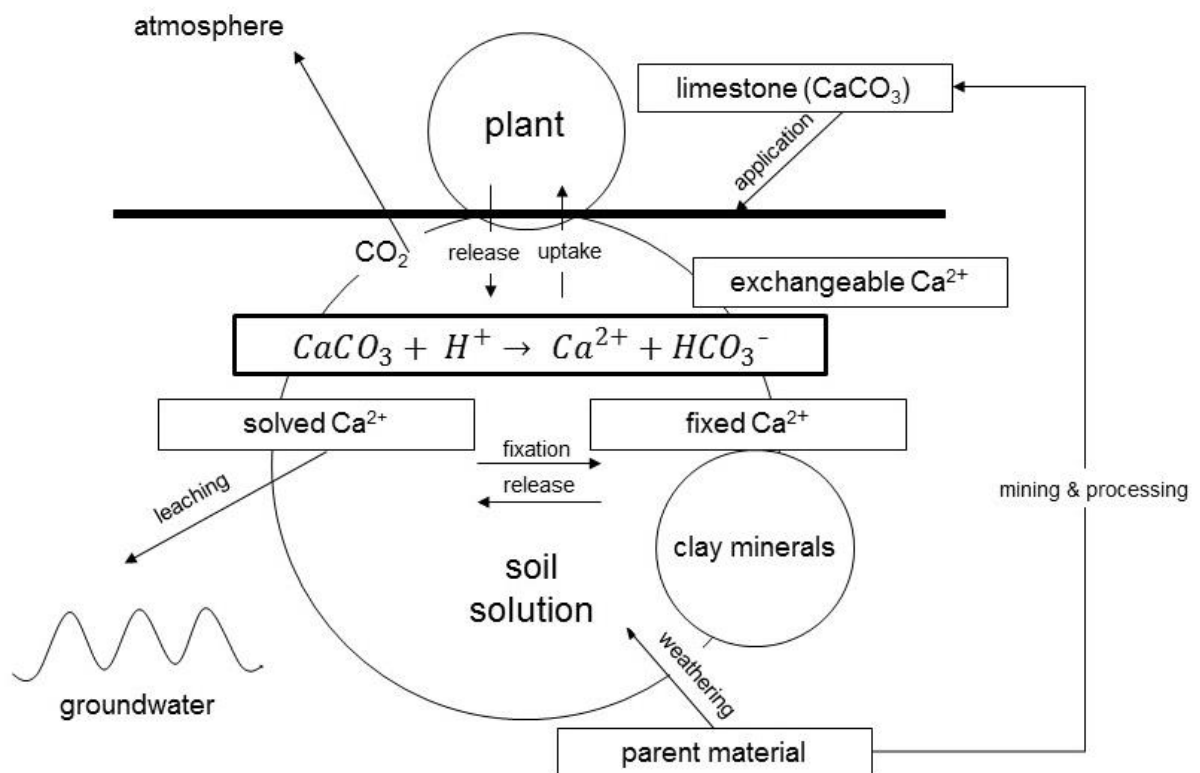


Fig. 3-2: The calcium cycle with lime application in soils and plants (redrawn and modified from Korb et al. (2005)).

The released Ca^{2+} further promotes the exchange and leaching of acidic cations (H^+ , Al^{3+} , Mn^{2+}) (Beetham, 2015; Holland et al., 2018). With the increasing pH value in the topsoil, the released positive Ca^{2+} ions are attracted to the negative clay surface and neutralize the charge in the clay, whereby the bonds can be completely covalent or ionic or a combination of both (Rengasamy et al., 2016). Negatively charged soil colloids (e.g. clay particles) will be surrounded by a diffuse double layer of cations in different stages of attraction to the colloid surface. The increase of the ionic force along

with the greater availability of soluble Ca^{2+} in the soil promotes a thinning of the diffuse double layer around soil colloids (Sparks, 2003). Consequently, the approach of individual clay particles is facilitated and they are more likely to join into flocs, enter face-to-face associations and form clusters (Dash and Hussain, 2015), or connect with negatively charged organic colloids with the help of calcium (Beetham, 2015; Haas et al., 2018b). Different models exist for the structure of this diffuse double layer. The double layer model by Stern (1924) divides the ion swarm around a charged colloid into an inner layer of counterions (Stern-layer) and an outer diffuse layer with counterions and co-ions (Sparks, 2003; Sposito, 1984). Due to the increased space required by the hydrated cations within this Stern-layer, the anion/cation ratio is larger. Thus, more anions are present than cations, resulting in a negative charge. To compensate this charge, the counterions are located in the diffuse layer, in which the cations and anions are subject to diffusion movement and no longer fixed. Due to the electrostatic attraction, there is a large excess of positive counterions near the particle surface, which decreases with increasing distance from the surface in favor of the anions. As a result, the potential approaches zero exponentially. Ideally, the surface charge in the liquid phase is completely compensated by ions and electron neutrality is established in the overall system. The diffusive motions release shear forces, which in some cases lead to reductions in the diffuse layer (e.g., during cation exchange). This results in a loss of particles of the diffuse layer and a lack of particles to compensate the Stern-layer (Baumgarten, 2013) and leads to a potential difference - the zeta potential ξ . The latter arises in the boundary region between the double layer of an exchange site and the surrounding solution, when the surrounding ion swarm of a particle is subjected to a shear movement (Heibach, 2018; Lagaly and Ziesmer, 2003).

The physical behavior of colloidal clay dispersions is essentially determined by the tendency of the particles to aggregate. As a result of Brownian molecular motion, collisions occur between particles dispersed in the liquid. The stability of the dispersion is determined by the interactions between the particles during the collision process. Attractive van der Waals forces on the one hand and repulsive forces, such as the repulsion of equally charged particles, on the other hand are responsible for the stability or aggregation behavior (Heibach, 2018). Derjaguin and Landau (1941) and

Verwey and Overbeek (1948) independently developed a theory on the stability of dispersions called the DLVO theory. They assumed the stability and aggregation behavior of dispersions as a consequence of energy changes when particles approach each other (Derjaguin et al., 1987). The formation of larger units of initially attached particles is called flocculation or coagulation resulting in a reduced ξ . The dispersed clay amount and flocculation values are related to net negative charge of clay particles and further depending on the clay mineral. Clay particles electrostatically attract positively charged ions such as Ca^{2+} , which form bridges holding the particles together. Therefore, these cations can induce flocculation of clay particles (Chibowski, 2011). The percentage increase in net negative charge with an increase in pH is in the order illite > kaolinite > smectite (Chorom and Rengasamy, 1995). Thus, especially clay and silt-clay soils (with a high amount of 3-layered clay minerals) are favored by treatments with lime, which are then typically characterized by flocculated particles in a “card-house” arrangement (Wuddivira and Camps-Roach, 2007). In a concept for describing the dispersive soil, Rengasamy et al. (2016) point to the reduction of dispersive charge or an increase in flocculation charge to restore dispersive soil. Liming with CaCO_3 releases Ca^{2+} ions in the soil that exchange other, more dispersive cations (Na^+), thereby reducing the dispersive charge and at the same time increasing the flocculation charge of the soil. The two primary reactions (i) cation exchange and (ii) flocculation take place rapidly in soils and reduce plasticity, improve workability and mechanical strength (Mallela et al., 2004) due to an increased aggregate stability. However, the exchange reactions do generally not affect the structure of clay particles themselves (Mitchell and Soga, 2005a). Consequently, the long-term stabilization in soil-lime mixtures increases with clay content (Bell, 1996; Maubec et al., 2017). These soil-lime mixtures lead to a well-connected pore system and stable aggregates (Chan and Heenan, 1999), which are mostly supported by the flocculation and agglomeration of clay and silt particles (Beetham, 2015; Bohne, 1983; Hartge and Ellies, 1977). The lime-induced soil rearrangement enhances the soil water storage capacity, saturated and unsaturated hydraulic conductivity, gas fluxes, organic matter and nutrient storage (Haynes and Naidu, 1998; Hoyt, 1981). The strengthening effect of lime on clay and loess soils (Bohne, 1983; Mordhorst et al., 2018) improves the susceptibility of the soil towards external stresses (Krümmelbein et al., 2013).

3.5 Objectives of this thesis

Liming is a well-established practice to ameliorate acidic soils worldwide. However, the role of liming in soil structure formation has only been little explored. It was hypothesized that improved physical soil properties and changes in aggregate stability are directly linked to soil structure. A better understanding is needed about the influence of soil management with lime, on the stability of macroaggregates and other physical parameters. Therefore, the present study deals with the effect of different levels of lime (CaCO_3) on various soil properties. Seven (six of them in this thesis) field trials were established with three different rates of lime in a randomized block design in various geomorphological units in Germany, with a wide range of different textures, clay contents and soil organic matter. Disturbed and undisturbed soil material was taken from the topsoil (0-30 cm) during the annual sampling (between the years 2017-2020) to quantify the stability or structural changes of the soil on the mesoscale (precompression stress and frame shear tests as well as crushing tests). Furthermore, the bulk density, pore size distribution, air- and saturated water conductivity were measured, and the chemical properties of the equilibrium soil solution were determined. The stability but also the structural change of a soil depends on the complex interaction and a wide range of soil properties and mechanisms. Such studies should be of a long-term nature, since lime distribution into the soil and soil aggregate formation by biological activities take time. The results are also helpful for a well-founded expert advice on the site-specific optimized lime supply of arable soils.

The main objectives of this thesis are:

(1.) to evaluate the relationship between different lime rates and the changes in the structure of arable soils via the determination of physical and mechanical parameters such as saturated hydraulic conductivity, water retention, shear strength and pre-compression stress values, and to reconsider the current lime recommendations for farming practices in Germany regarding the water availability as well as aspects of soil stabilization (Chapter 4: Frank, T., Zimmermann, I., Horn, R., 2019. The need for lime in dependence on clay content in arable crop production in Germany. *Soil and Tillage Research* 191, 11–17.),

(2.) to assess the shift of the pore size distribution, to estimate relevant pore functions (such as oxygen transport) and the improved infiltration during heavy rainfalls and water supply during dryer periods by an increased Ca^{2+} supply in soils with high clay content (Chapter 5: Frank, T., Zimmermann, I., Horn, R., 2020. Lime application in marshlands of Northern Germany – Influence of liming on the physicochemical and hydraulic properties of clayey soils. *Soil and Tillage Research* 204, 104730.),

(3.) to analyze the bulk density, and shear parameters as well as aggregate characteristics (tensile strength) under the influence of lime in different agricultural clayey soils (Chapter 6: Frank, T., Zimmermann, I., Horn, R., 2021. Effect of CaCO_3 application on mechanical strength and hydraulic properties in arable soils. *Soil Use and Management* (submitted)),

(4.) to determine changes of the erosive strength of aggregates and an improving accessibility of pores and particle surfaces after the application of lime (Chapter 7: Frank, T., Zimmermann, I., Horn, R., 2021. Impact of lime application on erosive strength and bulk density of aggregates. *International Agrophysics* (submitted)).

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PUBLICATIONS

4 The need of lime on clayey soils in arable crop production in Germany

Tina Frank, Iris Zimmermann, Rainer Horn (2019)

Soil and Tillage Research, Volume 191

4.1 Abstract

Liming is a well-established practice to ameliorate acidic soils worldwide. However, the role of liming in soil structure formation and its relation to the nitrogen cycling is only little explored. We hypothesized that improved physical and mechanical soil properties, which are directly linked to soil structure, lead to lower nitrogen losses in an agricultural system. Water holding capacity, total pore volume, bulk density, hydraulic conductivity, shear stress and pre-compression stress were quantified in the topsoil of three surveys located in Germany on different soil types: Eutric Gleyic Cambisol (Loamic) (Magdeburg, Saxony-Anhalt), Haplic Luvisol (Neubrandenburg, Mecklenburg-Western Pomerania) and Eutric Cambisol (Loamic) (Puch, Bavaria). The field experiments were arranged in a Randomized Complete Block Design and treated with two different rates of lime (calcium carbonate). Furthermore, three rates of nitrogen in form of urea or calcium ammonium nitrate were applied every year. Liming significantly increased the pH, plant available water capacity, total pore volume and decreased bulk density in the soils six months after application, but twelve months after lime application the soil structure collapsed again causes by ploughing and chiseling. We, therefore, assume a positive influence of lime application on soil physical parameters, if tillage is excluded at least initially. On the other hand, our results from the shear stress parameters and the pre-compression stress values indicate that their changes need time and we assume that it may take several years to develop a sustainable structure system.

4.2 Introduction

The degradation of arable soils due to natural acidification and the additional application of acidifying fertilizers is one of the factors endangering food security for a growing world population (Holland et al., 2018). Additionally, yields are directly affected by soil physical properties such as water retention capacity, bulk density, and hydraulic

conductivity. The persistence of soil properties is determined by soil structure formation and its strength, which also indicates that these properties are only valid within the rigidity limits (Horn et al., 2014). It is well known, that a disturbed soil structure with low water holding capacity, high bulk density, low saturated water conductivity as well as low soil strength will hamper crop growth (Finch et al.; Finch et al., 2014). Modern farming practices may cause growth problems and yield decline depending on climatic conditions and land use management due to higher and heavier machinery accompanied by higher mechanical stresses on the soils and risks of compaction (Horn et al., 2019). This compaction becomes irreversible when soils are subjected to mechanical stresses that exceed the elastic range of deformation and cause negative changes in the pore system (Arvidsson and Keller, 2004; Lebert and Horn, 1991). Therefore, the development of management practices that increase structural stability in arable soils is required. Water retention and hydraulic parameters, shear parameters such as cohesion and angle of internal friction as well as stress-strain data need to be quantified to assess the soil's resilience against mechanical impacts.

Lime application is a well-established practice for the amelioration of acidic soils caused by Al^{3+} and H^+ ions, to provide optimal conditions for plant growth (Kirkham et al., 2007). Furthermore, low soil pH enhances clay leaching under humid climates as well as clay dispersion and depletion in the topsoil weakens the soil structure and can reduce water infiltration if silting occurs at the soil surface. This can lead to intensified erosion, and due to the anaerobic conditions in the soil to denitrification, which causing losses of 15 to 30 % nitrogen (Ruhberg, 2017).

However, it is long known that liming can improve soil structure, and in this context, several studies and reviews have focused on the investigation of liming effects on soil physical and mechanical properties as indicators for an improved soil structure. Haynes and Naidu (1998) summarized the positive influences of lime application on soil physical conditions as a long-term effect in arable land. Holland et al. (2018) focused on the current lime conditions in the United Kingdom and outlined that physical processes due to liming are often not clear and have to be investigated further. Hellner et al. (2018) investigated limited effects of liming on macropores networks by means of X-ray tomography images on clayey soils. Bennett et al. (2014) concentrated on

long-term liming in Australia on acid and arid soils and determined an increasing aggregate stability and significantly higher hydraulic conductivity.

Nevertheless, already decades ago, Czeratzki (1972) described a direct improvement of soil tilth due to lime application in a clayey loam. Hartge and Ellies (1977) proved that lime application to heavy clay soils resulted in an improved water infiltration together with higher shear strength. Bohne (1983) documented that CaO or CaCO₃ application in loess and in clayey soils increased not only the shear parameters but also the hydraulic conductivity. Haynes and Naidu (1998) showed that lime supports the formation of a stable aggregate structure, particularly in heavy soils. Beetham (2015) described that clay particles are attracted closer to each other and form flocs by the addition of calcium ions which can be explained by cation exchange, its take place between the surfaces of the clay particles, often associated with metallic ions. The diffuse hydrous double layer which surrounds the clay particles is modified, clay structure alters and the potential of volume change reduces (Bell, 1996; Gay and Schad, 2000). The flocculation of clay particles causes an increase of intra-aggregate and inter-aggregate porosity (Beetham, 2015). Physical soil parameters will be influenced such as Hoyt (1981) determined that the addition of calcium carbonate on an acid Gray Luvisol and a Gleysol increased soil water holding capacity, which also led to increased yields of rapeseed. Castro et al. (1991) measured an improved water infiltration six months after lime application on three Oxisols under coffee plantations in Brazil. Most of the more recent studies on the effects of lime application on physical, chemical and economic soil parameters have been conducted in Oxisols (Brazil) and Alfisols (Australia). For example, Valzano et al. (2001) reported that liming can improve soil physical parameters such as infiltration, penetration resistance and water availability on a sodic red-brown earth in Australia. Bennett et al. (2014) detected an improved aggregate stability and increased hydraulic conductivity of a Red Sodosol in Australia. In Northern Europe, Blomquist et al. (2017) detected that CaO and Ca(OH)₂ created a finer tilth in the seedbed of arable soils (clay content 23-40 %) and had a positive effect on the aggregate stability. Consequently, further research is needed to determine whether the observed processes can be transferred to soils in Germany, and to integrate these results in site-specific practical liming recommendations.

In the past years, several authors discussed the sometimes unfavorable properties of arable soils in Germany, also against the background of lime balance and the declining amount of sold lime (Schnug and Haneklaus, 2002). Lausen (2012) stated that lime application is often neglected in agricultural land use. Currently, 58 % of the arable sandy soils in Schleswig-Holstein are undersupplied. Clayey and silty soil have a higher need of lime supply, because of their sorption and buffering capacity, but around 50 % of these soils are undersupplied, too. In Saxony-Anhalt, more than 25 % of arable soils are undersupplied and have an urgent need of lime (Stolpe and Schimpf, 2017). The derivation of lime requirements in Germany is based on the current soil pH (in CaCl_2), texture and organic matter content (VDLUFA, 2000a), but does not consider soil physical properties and functions.

Hence, the objectives of the presented study are:

- (i) to evaluate the relationship between different lime rates and the changes in the soil structure of arable soils via the determination of physical and mechanical parameters such as hydraulic water conductivity, water retention, shear strength and pre-compression stress values, and
- (ii) to reconsider the current lime recommendations for farming practices in Germany regarding to water availability as well as aspects of soil stabilization.

4.3 Material and methods

4.3.1 Experimental design

The study sites are located in Magdeburg (Saxony-Anhalt, Germany), Neubrandenburg (Mecklenburg-Western Pomerania, Germany) and Puch (Bavaria, Germany). The soils are classified according to IUSS Working Group WRB (2014) as Eutric Gleyic Cambisol (Loamic) at the study site in Magdeburg, Haplic Luvisol at the study site in Neubrandenburg and Eutric Cambisol (Loamic) at the study site in Puch. At each site, the test area was divided into 48 individual experimental plots to investigate the effect of different rates of lime on soil physical and mechanical parameters. Every plot had a size of 6 x 18 m, arranged in a Randomized Complete Block Design with four replicates per treatment, and the fields were managed with conventional farming including tillage every year (Table 4-1). Lime (CaCO_3) was

applied in two different rates (Lime1 = lime application according to site-specific recommendation and Lime1.5 = site-specific recommendation x 1.5) to achieve an optimum soil pH according to the soil specific recommendations by the Association of German Agricultural Analytic and Research Institutes (VDLUFA, 2000a) (Table 4-1). The derivation of lime requirements in Germany is based on the current soil pH (in CaCl₂), texture and organic matter content. The treatment Lime0 without lime additions serves as control treatment in our research. The lime was applied only once at the beginning of the experiment and immediately incorporated with a cultivator (Magdeburg: 5 cm depth; Neubrandenburg: 12 cm depth; Puch: 9 cm depth). Additionally, three rates of nitrogen (N) fertilizer (urea or calcium ammonium nitrate) were applied every year. The N-application rates were also based on the guidelines of VDLUFA (2010). The crops were sugar beet (2017) and winter wheat (2018) on the field trial in Magdeburg, maize (2017) and winter wheat (2018) on the field trial in Neubrandenburg and winter wheat (2017) and winter barley (2018) on the study site in Puch.

Table 4-1: Date of lime application, quantity of applied CaO-equivalents in dt ha⁻¹, pH recommendations (excerpts of recommendations by VDLUFA, 2000a), date of soil sampling, date of tillage in 2017 and 2018 and related depth.

	Magdeburg	Neubrandenburg	Puch
Date of lime application	04.04.2017	06.04.2017	24.10.2016
Lime1 in CaO dt ha ⁻¹	30	25	70
Lime1.5 in CaO dt ha ⁻¹	45	37.5	105
Recommended pH level	6.4–7.2	6.3–7.0	6.3–7.0
Soil Sampling	12.05/09.10.17 & 10.04.18	11.05/10.10.17 & 12.04.18	06.05/17.07.17 & 25.04.18
Tillage 2017	05.04. & 17.10.	03.05. & 16.10.	13.09. & 24.09.
Depth (cm)	20 cm	28 cm & 25 cm	23 cm & 9 cm
Tillage 2018	22.07. & 27.08.	19.10.	04.07.
Depth (cm)	8 cm & 20 cm	22 cm	5 cm

4.3.2 Soil sampling

Samples from the A horizons were collected six months and twelve months after lime application (except for Puch, where the soil was sampled ten and twenty months after lime application, because the field trial was established earlier) from the Lime0, Lime1 and Lime1.5 plots. Undisturbed soil cylinder cores, 5 cylinders of 236 cm³ and 100 cm³ each and one cylinder of 470 cm³ and disturbed soil samples were taken from 10-20 cm depth in each plot, summing up to 24 undisturbed and four disturbed samples per treatment.

4.3.3 Measurements on disturbed soil samples

For all three study sites chemical soil characteristics were analyzed on disturbed, air-dried, soil material sieved to ≤ 2 mm (n=4) (Table 4-2). Particle size distribution was determined by sieve and pipette technique according to Blume et al. (2011). Soil organic carbon content was analyzed according to DIN ISO 10694 (1995). The pH values were measured in CaCl₂ solution according to DIN ISO 11260 (2018) and total N was determined according to DIN ISO 13878 (1998).

Table 4-2: Organic carbon content, total N, Soil texture (IUSS Working Group WRB, 2014) of the A-horizons (0-30 cm) of untreated control plots at the three survey sites (n = 4).

	Neubrandenburg	Puch	Magdeburg
Organic carbon (OC) [%]	0.7	1.1	1.7
Total N [%]	0.07	0.13	0.17
Soil texture [%]			
Sand	64	58	26
Loam	25	28	42
Clay	11	14	32

4.3.4 Measurements on undisturbed cylinder samples

Water saturated soil samples (100 cm³) were used to determine saturated hydraulic conductivity (K_s) based on the hood permeameter method described by Hartge (1993). Soil samples obtained for pore size distribution analysis (100 cm³) were saturated with water, and subsequently stepwise drained using a vacuum plate (−6 kPa to −50 kPa),

and the parameters air capacity (AC, pores $\varnothing > 50 \mu\text{m}$) and plant available water capacity (PAW, pores $\varnothing 50 - 0.2 \mu\text{m}$) were derived from the corresponding volumetric water contents. To determine the fine pores (FP, $\varnothing < 0.2 \mu\text{m}$) and thus the non-plant available water, small cylinders (2 cm^3) were drained at air pressure of 1.5 MPa. Total pore volume (TP) is determined from the relationship between soil density and bulk density. The bulk density (ρ_B) is determined from undisturbed soil cores, dried at $105 \text{ }^\circ\text{C}$ for 24 h (Blume et al., 2011).

Shear strength was measured on undisturbed soil cores (236 m^3) at field capacity (-6 kPa) in a drained uniaxial confined compression test (direct shear test) at defined loads (20, 50, 100, 200 or 400 kPa). The shear strength parameters cohesion (c) and angle of internal friction (φ) are defined by the Mohr Coulomb failure line and result in:

$$\tau = \sigma_n \cdot \tan(\varphi) + c \quad \text{eq. 4-1}$$

where (φ) is the angle of internal friction, (c) the cohesion and (σ_n) the applied vertical stress (Kézdi and Rétháti, 1988).

Soil pre-compression stress (σ_{pc}) was derived from stress-strain measurements carried out under confined conditions at field capacity (-6 kPa), using a pneumatic multistep oedometer (uniaxial confined compression test) and five loads (20, 50, 100, 200, 400 kPa). Each load was applied for ten hours, followed by three hours of relief. σ_{pc} values were determined graphically following the method of Casagrande (1936).

4.3.5 Data analysis

Data analysis and statistics were performed in R studio version 3.5.1 (R Core Team, 2017). The effects of the three lime rates on the different soil properties were evaluated. It was assumed that the data are normally distributed and have homogeneity of variance. Data of K_s , which are not normally distributed, were logarithmized. The data were tested by applying analyses of variance (One-Factor ANOVA) for one factor of interest (Snedecor and Cochran, 1996). All values were evaluated comparing the means of the factor levels treatment and plot for each site separately. The significance of the different tests was set at an α -Level of 5 %.

4.4 Results

In Magdeburg, with a clay content of 32 %, the pH slightly increased with increasing lime application for all plots (Table 4-3). Twelve months after the lime application, the pH slightly decreased. The average ρ_B decreased on the limed plots by between 12 and 14.5 % relatively to the control six months after lime application. In contrast, TP, AC as well as PAW were significantly lower in the Lime0 plots than in the Lime1 plots ($p < 0.05$) six months after the lime incorporation. FP were significantly lower for Lime1 plots compared to Lime0 ($p < 0.05$) six months after lime application. Lime1.5 treatment resulted in less changed retention parameters than for Lime1 (optimal level) treatment (Table 4-3).

Table 4-3: Average pH in CaCl_2 ($n = 4$), ρ_B and water retention parameters: Air capacity (AC), plant available water capacity (PAW), fine pores (FP) and total pore volume (TP) ($n = 8$) in the 10-20 cm depth at the study site in Magdeburg, six (2017) and twelve (2018) months after lime application and incorporation. * indicates treatment significantly different from control (Lime0) at $p < 0.05$.

	pH (CaCl_2)	ρ_B g cm^{-3}	AC Vol.-%	PAW Vol.-%	FP Vol.-%	TP Vol.-%
Sampling 6 months after lime application						
Lime0	6.1	1.46	11	6	27	45
Lime1	7.1	1.25	18*	12*	23*	53*
Lime1.5	7.1	1.28	16	9	27	51
Sampling 12 months after lime application						
Lime0	6.4	1.53	2	4	38	43
Lime1	6.7	1.56	5	8*	30	42
Lime1.5	6.7	1.61	4	6	34	43

Lime0 (no lime application), Lime1 (lime application according to site-specific recommendation), Lime1.5 (site-specific recommendation x 1.5)

In Neubrandenburg, with a clay content of 11 %, the pH increased six months after lime application and was almost stable twelve months after lime application (Table 4-4). Six months after lime incorporation, the average ρ_B in the limed plots Lime1 and Lime1.5 was between 4 and 7 %, respectively lower than in the Lime0 treatment. Only the FP of Lime1 plots were significantly affected ($p < 0.05$) and decreased six months after lime application.

Table 4-4: Average pH in CaCl_2 ($n = 4$), ρ_B and water retention parameters: Air capacity (AC), plant available water capacity (PAW), fine pores (FP) and total pore volume (TP) ($n = 8$) in the 10-20 cm depth at the study site in Neubrandenburg, six (resp. ten) (2017) and twelve (resp. twenty) (2018) months after lime application and incorporation. * indicates treatment significantly different from control (Lime0) at $p < 0.05$.

	pH (CaCl_2) -	ρ_B g cm ⁻³	AC Vol.-%	PAW Vol.-%	FP Vol.-%	TP Vol.-%
Sampling 6 months after lime application						
Lime0	5.9	1.67	11	9	17	37
Lime1	6.8	1.56	14	15*	11*	41*
Lime1.5	6.9	1.59	13	18*	18	40
Sampling 12 months after lime application						
Lime0	6.2	1.66	7	15	14	37
Lime1	6.8	1.68	7	14	16	36
Lime1.5	6.9	1.63	9	12*	17	38

Lime0 (no lime application), Lime1 (lime application according to site-specific recommendation), Lime1.5 (site-specific recommendation x 1.5)

In Puch, with a clay content of 15 %, the pH slightly increased on the limed plots ten months after lime application (Table 4-5). Twenty months after the lime application, the pH slightly decreased. Liming did not distinctly affect the ρ_B or the TP at this site, neither ten nor twenty months after the lime incorporation. However, PAW significantly increased in Lime1.5 plots twenty months after lime application ($p < 0.05$).

Table 4-5: Average pH in CaCl₂ (n = 4), ρ_B and water retention parameters: Air capacity (AC), plant available water capacity (PAW), fine pores (FP) and total pore volume (TP) (n = 8) in the 10-20 cm depth at the study site in Puch, ten (2017) and twenty (2018) months after lime application and incorporation. * indicates treatment significantly different from control (Lime0) at p < 0.05.

	pH (CaCl ₂)	ρ_B g cm ⁻³	AC Vol.-%	PAW Vol.-%	FP Vol.-%	TP Vol.-%
Sampling 10 months after lime application						
Lime0	5.3	1.47	14	5	25	44
Lime1	6.8	1.52	11	9	23	42
Lime1.5	7.0	1.54	9	9	24	41
Sampling 20 months after lime application						
Lime0	5.6	1.50	10	15	19	43
Lime1	6.3	1.49	10	17	16	43
Lime1.5	6.1	1.53	8	22*	12*	42

Lime0 (no lime application), Lime1 (lime application according to site-specific recommendation), Lime1.5 (site-specific recommendation x 1.5)

Liming significantly increased K_s (p < 0.05) compared to the control plots (Lime0) six months (resp. ten months) after the lime application in all three field trials. The results are classified according to Ad-hoc-AG Boden (2005) as very low (< 1 cm d⁻¹) to very high (100 to < 300 cm d⁻¹) values and to simply, results are shown in logarithmic display (Fig. 4-1). In Magdeburg, K_s increased on Lime1 plots and Lime1.5 plots in the first six months after the lime application. In Neubrandenburg, K_s increased on Lime1 plots and on Lime1.5 plots in the first six months after the lime application. In Puch, K_s increased on Lime1 plots and Lime1.5 plots in the first ten months after the lime application. Twenty months after the lime application, no significant differences between the three treatments were detected on the study site in Puch. In Neubrandenburg, K_s is significantly higher in Lime1.5 (p < 0.05) plots than on the Lime0 and Lime1 plots, twelve months after the lime application. In Magdeburg, K_s is significantly higher on Lime1 (p < 0.05) plots than in the Lime0 plots twelve months after lime application.

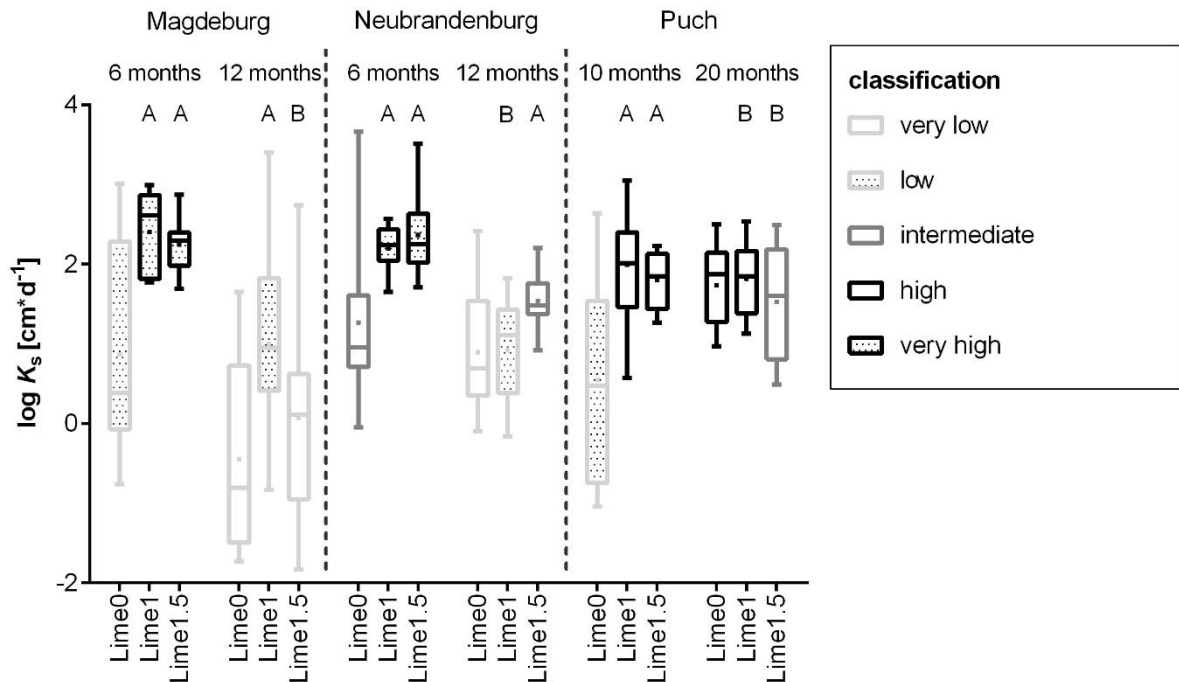


Fig. 4-1: Saturated hydraulic conductivity (K_s) (logarithmic display) in the 10-20 cm depth at the three survey sites Magdeburg, Neubrandenburg and Puch, six (resp. ten) (2017) and twelve (resp. twenty) (2018) months after lime application and incorporation. Classification of K_s according to Ad-hoc-AG Boden (2005), $n = 12$. A (B) indicates treatment (not) significantly different from the control (Lime0) at $p < 0.05$.

Internal strength of the soil was determined by means of the pre-compression stress, σ_{pc} , for all plots from all three field trials (Fig. 4-2). In the topsoil, σ_{pc} did not differ significantly among the three field trials six months (resp. ten months in Puch) as well as twelve months (resp. twenty months in Puch) after lime application. Mostly, the treatments show a broad distribution of σ_{pc} from 12 to 78 kPa classified as low to medium values. The vertical line at 60 kPa in Fig. 4-2 indicates the shift from low to medium values (DVWK, 1995b) (Table 4-6).

Table 4-6: Classification of the mechanical pre-compression stress σ_{pc} [kPa] according to (DVWK, 1995b)

level	classification	pre-compression stress σ_{pc} [kPa]
very low	1	≤ 30
low	2	30-60
medium	3	60-90
high	4	90-120
very high	5	120-150
extremely high	6	≥ 150

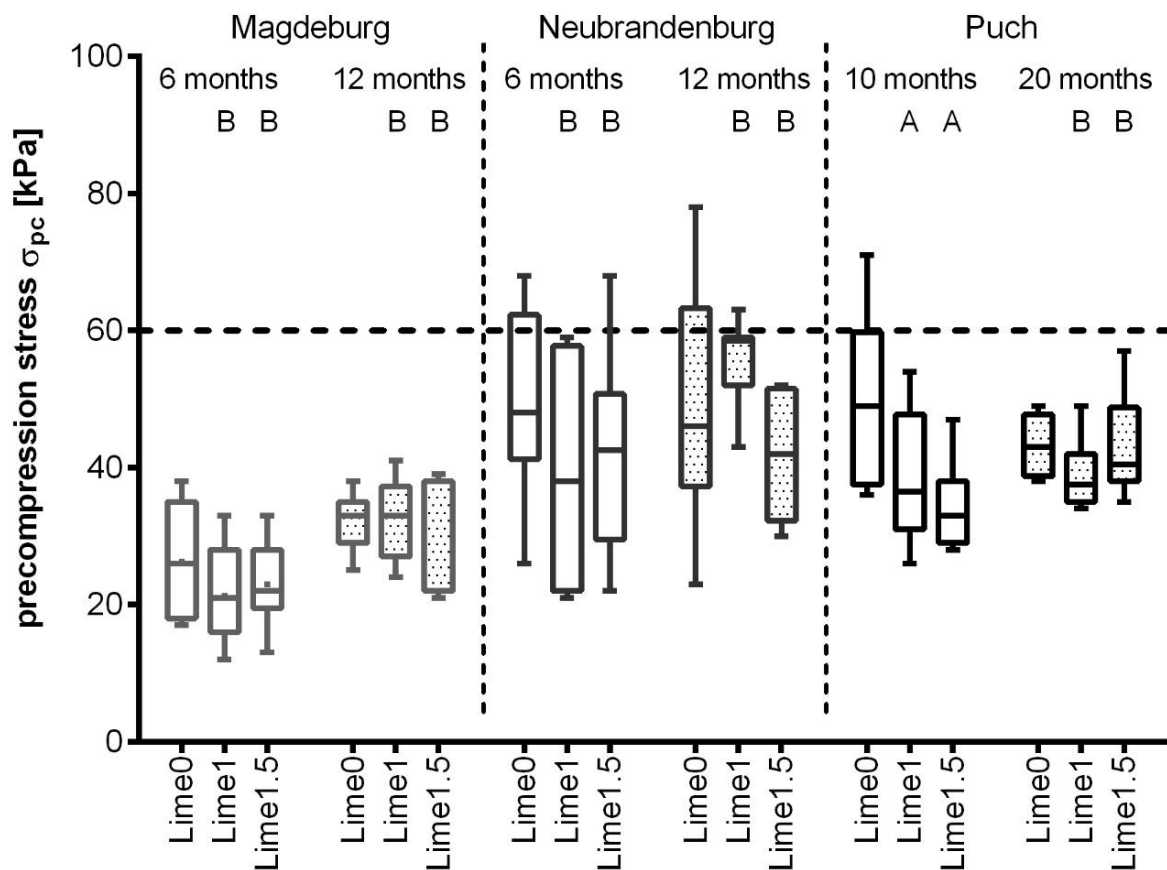


Fig. 4-2: Pre-compression stress (σ_{pc}) at a soil matric potential of -6 kPa in the 10-20 cm depth at the three survey sites Magdeburg, Neubrandenburg and Puch, six (resp. ten) (2017) and twelve (resp. twenty) (2018) months after lime application and incorporation, $n = 8$. A (B) indicates treatment (not) significantly different from the control (Lime0) at $p < 0.05$.

Similar to the results obtained for σ_{pc} , no effects of liming were observed on shear stress parameters (Table 4-7). However, tendencies to a higher c in Lime1 treatments are visible in Neubrandenburg six months after lime application and in Magdeburg twelve months after lime application. In Puch are neither after ten months nor after twenty months any measurable changes to be seen for the shear parameters.

Table 4-7: Cohesion (c) in kPa and angle of internal friction (φ) ($n = 4$) in deg. (-6 kPa) in the 10-20 cm depth at the three survey sites Magdeburg, Neubrandenburg and Puch six (resp. ten) (2017) and twelve (resp. twenty) (2018) months after lime application and incorporation.

	Magdeburg		Neubrandenburg		Puch	
	c kPa	φ deg.	c kPa	φ deg.	c kPa	φ deg.
	Sampling 6 (10) months after lime application					
Lime0	15.8	33.8	11.5	38.8	25.5	33.9
Lime1	11.2	33.5	24.5	31.2	13.9	36.8
Lime1.5	16.8	33.9	12.8	39.2	23.3	30.5
	Sampling 12 (20) months after lime application					
Lime0	23.8	34.3	19.1	36.6	21.0	32.6
Lime1	32.0	31.7	22.2	33.8	18.2	36.0
Lime1.5	18.6	32.7	22.7	34.9	23.9	33.6

Lime0 (no lime application), Lime1 (lime application according to site-specific recommendation), Lime1.5 (site-specific recommendation x 1.5)

4.5 Discussion

The current discussion between farmers and advisory boards in collaboration with scientists concerning the optimal soil structure and highest and sustaining accessibility of mineral particle surface properties and pore functions is also often related to chemical interactions. The question how far are the actual recommendations of e.g. liming still valid or do we need to alter the recommended lime application amount in this context is often posed amongst others by Hartge and Bohne (1983) and Hartge and Bohne (1985). However, since then the debate about the optimal pH and CaCO_3 content especially for loamy soils are controversial (VDLUFA, 2000a) and seem to not include the documented strengthening effect of cations like Ca^{2+} even on the microscale (Holthusen et al., 2011) and the positive interactions concerning hydraulic

functions. Our results in general support the positive interactions and effects on physical properties at the three field trials with substantial variation in soil texture and clay content. Generally, we found significant differences between the three treatments (no liming, recommended lime amount and 1.5 times the recommended lime amount) mostly for the retention parameters and the saturated hydraulic water conductivity. While no clear interactions between lime application and mechanical parameters were observed throughout the whole period, since the lime-induced changes of σ_{pc} , ϕ and c were mostly not significant. If we compare our results with Bohne et al. (1985) for undisturbed soil, these effects were to be foreseen because the annual tillage, carried out six months before taking the soil samples, in the first 30 cm of the topsoil, and homogenized the newly commenced structure formation (Kuncoro et al., 2014; Schjønning et al., 2015).

Obviously, liming increased the soil pH of all three trials. Thereby the target pH-value according to VDLUFA (2000a) was reached at all three sites. However, this measurable positive effect requires a more detailed analysis concerning its relation to soil functions. The pH analysis defines an average value of homogenized samples and does not indicate actual in-situ conditions. For example, as rainfall increases, H^+ ions predominate the soil solution and promote acidification (Carver and Ownby, 1995). Reth et al. (2005) stated that extreme climate conditions would indirectly affect the soil pH, while they directly affect the nutrient cycle and plant available water. In both sampling years, extreme conditions occurred with high rainfalls in autumn 2017, contrary to the rainfalls the years before (enhanced leaching of basic cations) which possibly affected our in-situ pH values and altered the hydraulic functions due to structure collapse which is also in agreement with the findings of Hartmann et al. (1998). Intense shrinkage in the extremely dry summer 2018 affected the water storage as well as the hydraulic functions. Moreover, nutrient uptake by plants affect the ion composition in the rhizosphere and changes the soil strength. Finally, also the fertilizer effects need to be included in the whole discussion. Ammonium based N-fertilizers such as calcium ammonium nitrate, used in our field trials, have the greatest potential to acidify soils because they generate two H^+ ions for each ammonium molecule nitrified to nitrate. The extent of acidification depends on whether the nitrate produced

from ammonium is leached or is taken up by plants (Bouman et al., 1995). It is possible that the used fertilizers supported the acidification process in our research.

The application of lime generally reduced ρ_B (six months after lime application) in our field trials. Anikwe et al. (2016) observed similar results for the application of lime and gypsum on a degraded Ultisol in a non-tillage system. Furthermore on silty and clayey soils in Brazil, Auler et al. (2017b) supported our initial findings as they determined an increased total porosity and a shift in the pore size distribution in topsoil layers after the addition of lime. But, they did not describe a decrease because they applied lime on the surface without subsequent tillage. However, after tillage we detected no differences in the ρ_B between the lime treatments twelve months after lime application. Actually, tillage leads to lower ρ_B due to soil loosening, and any additional load on the soil by machines compacts the soil (Blume et al., 2016b). Urbanek et al. (2014) showed that tillage has a negative effect on the strength of different aggregate sizes. During the tillage operations, the actual soil structure will be destroyed and the next crack formation during drying starts again, resulting in macropores between the weakened aggregates. Consequently, the restructuring requires time and lime application can only support and strengthen these newly formed aggregates (Beese et al., 1979; Hartge and Ellies, 1977) as can be assumed from our results six months after lime application. If during the following tillage these slightly strengthened aggregates are destroyed again, the lime effect is also weakened, and lime could not permanently stabilize the soil structure in order to prevent harmful influences for soil pores system.

We consider the changes in the pore size distribution and combine them with the possible strengthening and changes in the accessibility of pore surfaces or pore continuity on the long-term. We may take the shift in the pore size distribution from fine pores towards narrow coarse pores and medium pores on limed plots as an initial hint for positive structure formation processes, caused by the effect that calcium promotes flocculation. This flocculation of clay particles, and the increased soil resistance to compression are already mentioned by Hartge (1959a) and Haynes and Naidu (1998). They stated that after a period of 6 months, liming resulted in improved soil aggregation. In addition to the pore size distribution, the soil stability plays an essential role. However, such strengthening takes time because the rearrangement of particles during the formation of new aggregates always requires a replacement at the available

particle surfaces or the mobilization of shear resistance at a given surface (Horn and Lebert, 1994). Thus, our results prove that the initial increase in soil strength is very vulnerable and collapses if external forces (tillage) are applied.

As we already stated are the water retention parameters negatively affected presumable due to tillage on some sites, even if we found for the field trial in Neubrandenburg on Lime1 plots, six months after lime application a positive relationship between plant available water (PAW) and addition of lime. Nonetheless, based on the presented results and existing literature, we assume a positive influence of lime application on the soil structure, visible in a shift of the pore size distribution, which affected PAW and TP as long as the structure formation is not interrupted by a following tillage.

It is hypothesized that applied lime can support the development of a pore network with well-connected pores. A long-term experiment conducted on a Red Sodosol in Australia by Bennett et al. (2014) determined significant higher K_s values, and an improved pore network with major preferential flow paths in the lime treatments. Wuddivira and Camps-Roach (2007) argued that calcium and organic matter support the formation and stability of soil aggregates and thereby increase K_s in clay-rich soils. Yilmaz et al. (2005) reported a positive relationship between increasing K_s and addition of lime as a result of the flocculation of clay particles in the presence of Ca^{2+} ions. Correspondingly, in our results, K_s increased significantly with the application and incorporation of lime on clayey arable soils. Kirkham et al. (2007) observed that liming doubled the saturated hydraulic conductivity in a long term experiment in Australia and explained this result with an increase in medium and coarser pores. In the present study, we could connect the higher K_s values to an increase of narrow coarse pores and medium pores, even twelve months after the lime application. The consequences of an appropriate lime application amongst others are an improved water flow in the topsoil, as it was found in the limed plots in our field trials six to ten months after lime application. The extent to which such positive effects can be expected depends however on the texture as well as the sensitivity to soil aggregate collapse if external forces like tillage are applied.

Especially the soil in Magdeburg is very sensitive to further soil degradation as can be seen by the decreasing K_s values decreased even after the application of higher rates of lime application due to tillage.

In general, the creation of larger pores not necessarily increase the hydraulic conductivity, rather, the pore-connectivity also needs to be increased. It is important to know, if there is an improvement in soil physical properties, which is reflected in soil parameters for example the link between the pore size distribution and bulk density with soil strength (Lebert and Horn, 1991). In the present study, we found differences in σ_{pc} between the three lime treatments on all field trials, though non-significant. Hence, our results correspond to findings from other authors and support that the pre-compression stress values are highly vulnerable to soil tillage (Arvidsson and Keller, 2004). In most present studies, where lime was not subjected to tillage, higher σ_{pc} values were obtained for soil-lime mixtures (Locat et al., 1990; Pereira et al., 2018).

Because of the short-term restructuring followed by the again deterioration during tillage have the lime treatments in our survey to date not notably affected the shear parameters c and ϕ in the soils. Thus, soil tillage is counterbalancing the positive effects of the lime application on mechanical parameters. We can at least detect the beginning of an improved structure formation and an increased strength as the cohesion values increased within the first time interval between lime application and sampling (six months) which was also reported by Gay and Schad (2000) after addition of quicklime on a silty soil (10 % clay) without soil tillage. Thus, the current study dealing with soil mechanical parameters has emphasized that the close interactions between lime addition on clayey soils and the effects on soil strength need to be taken into account in lime management even if more research is needed to completely understand the mechanism of action. A tendency of a change in the soil structure with time after the addition of lime to arable soils can be finally also derived from our results. Consequently, we also need to address the kind of lime application to get a better result of higher pH as well as nutrient sorption and accessibility in combination with the formation of stronger aggregates, which develop a sustainable structure and change soil mechanical properties via liming over several years.

4.6 Conclusions

- Although liming had only little effect on soil mechanical parameters when the topsoil's underwent conventional tillage practice, significant changes in soil physical parameters were determined six months after lime incorporation on the three field trials (without being interrupted by tillage). The stabilization process of the soil by liming is reflected in a shift of pore size distribution from fine pores towards narrow coarse pores and medium pores six months after lime application. The disruptive effect of tillage is immediately visible in lower plant available water twelve months after lime application (with tillage in between) on all plots. In general, tillage interferes with the development of structure and functions in limed soils.
- Because the physical resilience of a soil is related to its structural stability, we need to use a site-specific tillage or land use management to preserve and further strengthen soil structure.
- Further research should focus on the one hand how the lime-induced improvement of mechanical parameters related to soil structure can affect important soil fertility factors, such as e.g. the N cycle in arable land. Secondly, research should concentrate on the effect of tillage on limed soil systems, in shorter sampling time intervals to detect temporal changes and to better quantify the tillage effect. After that, it will be possible to reconsider the current lime recommendations.

4.7 References

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5 Lime application in marshlands of Northern Germany – Influence of liming on the physicochemical and hydraulic properties of clayey soils

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5.1 Abstract

Liming is a well-established practice to ameliorate acidic soils worldwide. However, 42 % of the arable soils in Germany are classified in a low pH-range (A/B). We hypothesize that the surface application and incorporation of lime will affect the structure and thus the physicochemical and hydraulic properties of two marshland soils with different clay content. Water holding capacity, total porosity, bulk density, hydraulic conductivity, and aeration parameters were quantified in the topsoil of two study sites located in the marshlands of Schleswig-Holstein, Germany. The soil at the first study site is a Haplic Gleysol, the most representative soil type in the region, with a clay content of 28 %, and the second is a Haplic Gleysol with a clay content of 40 %. The field experiments were arranged in a randomized block design with three different treatments: a control with no lime application (Lime0), lime application according to the soil-specific recommendations by (VDLUFA, 2000a) to achieve an optimized soil pH (Lime1), and soil-specific recommendations x 1.5 (Lime1.5). Liming significantly increased the pH, cation exchange capacity, plant available water capacity, and aeration parameters, decreased bulk density in the soils already 12 months after application, and the effects remained stable during the surveyed three years after lime application. The results underline the importance of sufficient lime supply and the potentials of elevated lime doses to ameliorate soil structure, plant water supply and yields in undersupplied, clay-rich soils.

5.2 Introduction

Climate change has a growing effect on soil and agricultural productivity (Rengel, 2011) as it requests soils with high fertility, excellent accessibility of particle and pore

surfaces as well as a sufficient pore connectivity down to depth to ensure yields (Horn and Blum, 2020). Increased leaching or surface runoff in periods with intense rainfall not only counteract plant growth and yield but also promote structure degradation and coinciding water ponding, flooded fields, surface runoff (Finch et al., 2014) and erosion. At present, forty-two percent of the arable soils in Germany are classified in a low pH-range (A/B) according to the association of German Agricultural Analytic and Research Institutes (VDLUFA, 2000a), with a poor soil structure, reduced nutrient availability, mobilization and increased plant availability of heavy metals, and consequently serious yield losses. In these soils, lime application is the most important fertilization measure to restore the productivity, but also to protect the soil against erosion, capping, and compaction (Jacobs et al., 2018). However, even if soils presently are at an optimal pH level (C) with a good soil structure and availability of nutrients, it is still important to support them with regular lime applications to keep them in the optimal pH-range, because annually around 400-600 kg/ha CaO is either taken up by roots, leached out by percolating infiltration water or consumed in the compensation of acidification (Rechsteiner and Keller, 2019). Thus, an application of lime on arable soils is necessary at least every three years, as it is recommended by VDLUFA (2000a). In the marshlands, used for agricultural production in Schleswig-Holstein, around 50 % of the soils are undersupplied with lime (Lausen, 2012), and this causes intensive slaking, and structure degradation with coinciding problems of restricted water availability, aeration and root growth.

The geological development of the marshlands and the origin of the natural environment depict the very special soil development in the shoreline region, because up to date it is primarily affected by the sea level rising during the Holocene and the daily tidal effects. The term “chronosequence” is used to demonstrate the degree of soil development under a varying duration of soil formation. Coastal salt marsh soils (IUSS Working Group WRB, 2014), termed “Rohmarsch” according to the German soil classification system (Ad-hoc-AG Boden, 2005), are defined as the initial state of the marsh soils chronosequence. In this region, dikes protected the land already in the 13th century in order to avoid flooding (Behre, 2004). The salt marsh areas are located in front of the dikes towards the sea (Spohn et al., 2013), while behind the dikes, calcareous marsh soils, termed “Kalkmarsch” according to Ad-hoc-AG Boden (2005)

or Calcaric Gleysols according to IUSS Working Group WRB (2014), dominate most of the younger polders at the North Sea coast in Schleswig-Holstein and are characterized by a high amount of calcium carbonates in the top- and subsoil, leading to high structural stability (Burbaum, 2008). They are mainly used as arable land because of their high productivity. However, acidic precipitation (Goulding, 2016), nutrient uptake by crops and roots (Hinsinger et al., 2003) and the application of acidifying fertilizers (Felix-Henningsen et al., 1983; Goulding, 2016) lead to a continuous decalcification of the marsh soils at rates of approx. 1 dm/100 yr. (Burbaum, 2008). The advanced weathering and formation of secondary minerals often lead to high clay contents in the decalcified marsh soils, which are termed Haplic Gleysols according to IUSS Working Group WRB (2014), and “Kleimarsch” according to Ad-hoc-AG Boden (2005). Because of the resulting short time slots of trafficability during the field season and the high risk of irreversible compaction, these soils are often described as “minute soils”.

The application of lime is well established in agricultural management, mostly in the form of limestone (CaCO_3) (VDLUFA, 2000a), and in civil engineering for earthwork projects in the form of quicklime (CaO) (Gay and Schad, 2000). Quicklime readily dissolves in the soil. The released Ca^{2+} promotes the arrangement of clay particles in stable associations and thereby helps the fast development of a porous and stabilized soil structure (Metelková et al., 2012; Mordhorst et al., 2018; Zimmermann et al., 2016). Limestone dissolves much slower than quicklime (Keiblinger et al., 2016), and this can decrease or retard the strengthening effect in soils (Frank et al., 2019). Positive impacts of quicklime, limestone or mixed lime applications on soil hydraulic characteristics, nutrient availability, and yields have frequently been found in European field experiments. Ulén and Etana (2014) demonstrated reduced phosphorus leaching due to the application of quicklime and slaked lime (CaO and Ca(OH)_2) in two clayey soils, which were dominated by the 2:1 mineral illite. The application of slaked lime and a mixed product of calcium carbonate and slaked lime can significantly increase aggregate stability and modify the aggregate size distribution in the topsoil for a finer seedbed (Blomquist et al., 2017).

In Germany, research on field trials that considers the influence of lime applications on soil physical properties is rare, especially in recent years. Field experiments by Hartge (1959a, 1959b) proved that liming increases the structural stability of arable soils. Laboratory experiments by Kuntze (1983) revealed that high limestone rates increase the water content at the liquid limit, and can stabilize marsh soils at high matric potentials. Rogasik et al. (2005) discovered an increasing base saturation with treatments of limestone in a Luvisol. They concluded that investigations on the cation exchange capacity in the soil are necessary to determine recommendations for lime application rates.

The marsh soils in Schleswig-Holstein are characterized by the interaction of swelling and shrinkage, promoted by i.e. 2:1 clay minerals like smectites. Swelling can change the pore size distribution towards finer pores, so the infiltration of water is no longer assured (Beetham, 2015; Haynes and Naidu, 1998). It is well known, that lime application can limit swelling, which is partly due to the suppressive effect of Ca^{2+} ions in the diffuse double layer (Ulén and Etana, 2014). Ca^{2+} ions in the soil solution can reline the surfaces of the clay particles and promote the displacement dispersive cations e.g. Na^+ . This enhances flocculation and agglomeration, and affects physical characteristics in soils, such as the pore size distribution or water infiltration (Beetham, 2015; Bell, 1996; Choquette et al., 1987; Hartge and Ellies, 1977). When the diffuse double layer surrounding the clay particles is compressed, clay particles are more likely to join into flocs (Beetham, 2015; Gay and Schad, 2000; Reeves et al., 2006), get in face-to-face associations and form clusters (Dash and Hussain, 2015). This leads to the development of a “card house structure” with increased inter- and intra-aggregate porosity (Holthusen, 2010; Muneer and Oades, 1989b; Oades, 1984a).

In combination with a reduced soil strength, the repeated wheel traffic with increasingly heavier machines causes compaction and consequently a loss of pore volume, connectivity and functionality, especially under moist and wet soil conditions (Blume and Pfisterer, 1993; Duttmann et al., 2014; Horn et al., 2019). As the flow of water and air in soils largely depends on the continuity of the pore network, changes in the pore system can be detected by hydraulic conductivity and air permeability measurements as a function of matric potential (Ball et al., 1988; Hamamoto et al., 2011).

The present derivation of lime requirements in Germany is based on the current soil pH, texture and organic matter content (VDLUFA, 2000a), but physical properties are not considered. Therefore, the effects of lime applications on the hydraulic, pneumatic properties and functions of arable soils are described in this work. Our key research questions are:

- Can the increased Ca^{2+} supply support a shift of the pore size distribution to fewer fine pores and a higher plant available water capacity?
- Can limestone applications help to improve infiltration during heavy rainfalls and water supply during dryer periods?
- Is the amelioration of relevant pore functions, especially for oxygen transport, more intense in soils with higher clay content, and can this be verified by aeration properties?

5.3 Material and methods

5.3.1 Experimental design and soil sampling

Two study sites were established on arable land in summer 2016. The field experiments included three treatments: A control with no lime application (Lime0), lime application according to the soil-specific recommendations by (VDLUFA, 2000a) to achieve an optimized soil pH (Lime1), and soil-specific recommendations x 1.5 (Lime1.5). The applied fine-grained limestone contains low amounts of magnesium and the neutralizing value is 48 %. The treatments were arranged in a randomized block design with four replicates. Every plot had a size of 6 x 18 m, and local farmers managed the fields conventionally with annual tillage. The sites are located in the western part of Schleswig-Holstein close to the North Sea in Germany (Fig. 5-1).

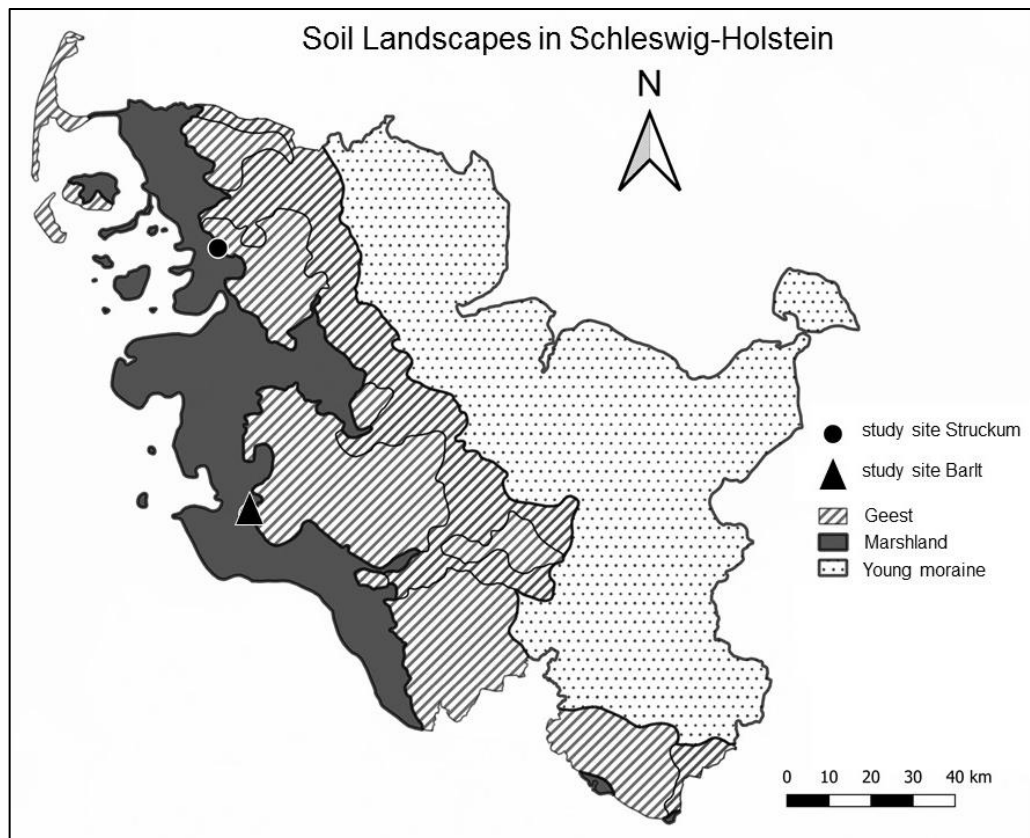


Fig. 5-1: Map of the different soil landscapes in Schleswig-Holstein and the study sites Bartl and Struckum. Bundesamt für Kartographie und Geodäsie (BKG) (2010). Vector data of the digital landscape model 1:250.000.

The site “Struckum” is in the district of North Friesland and the soil classified as a Haplic Gleysol according to IUSS Working Group WRB (2014) and a “Kleimarsch” according to Ad-hoc-AG Boden (2005). The topsoil is a silty clay loam with a clay content of 28 % and an organic carbon content of 9 g kg^{-1} (Table 5-1). The dominant clay mineral is the 2:1 mineral illite. The start-pH is 5.0 and the recommended pH is between 6.4 and 7.2 (VDLUFA, 2000a). On this site, the lime amount corresponding to lime recommendation according to VDLUFA (2000a) x 1.5 exceeded the permitted annual lime application amount for Schleswig-Holstein. Consequently, the Lime1.5 treatment on this site is equivalent to the lime recommendation according to VDLUFA (2000a), while the Lime1 treatment represents 2/3 of the amount according to the lime recommendation by VDLUFA (2000a). The lime application rates were $80 \text{ dt CaO ha}^{-1}$ in the “Lime1”-treatment and $120 \text{ dt CaO ha}^{-1}$ in the “Lime1.5”-treatment. The crops were winter wheat (2017), oats (2018) and winter rape (2019).

The second field trial “Barlt” is located around 80 km south of Struckum in the district Dithmarschen. The soil is a Haplic Gleysol according to the IUSS Working Group WRB (2014) and a “Kleimarsch” according to Ad-hoc-AG Boden (2005). The topsoil is a silty clay with a clay content of 45 % and an organic carbon content of 56 g kg⁻¹ (Table 5-1). The dominant clay minerals are the 2:1 minerals montmorillonite and illite. The start-pH is 5.9 and the recommended pH is between 5.9 and 6.7 (VDLUFA, 2000a). The lime application rates were 36 dt CaO ha⁻¹ in the “Lime1”-treatment and 54 dt CaO ha⁻¹ in the “Lime1.5”-treatment. The crops were winter wheat (2017), wheat (2018) and winter wheat (2019). On both study sites, the lime was applied and incorporated with a plough (~ 25 cm) only once at the beginning of the field trials in 2016 before sowing. Soil sampling took place 12 (2017), 24 (2018) and 32 (2019) months after lime application from the topsoil of the Lime0, Lime1 and Lime1.5 plots (Table 5-2).

Table 5-1: Organic carbon content, soil particle size (IUSS Working Group WRB, 2014) of the A-horizons (0–30 cm) of untreated control plots at the two sampling sites (n = 4).

	Struckum	Barlt
Organic carbon [g kg ⁻¹]	9.0	56.0
Sand [%]	9.0	5.0
Silt [%]	63.0	50.0
Clay [%]	28.0	45.0

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Table 5-2: Date of lime application, quantity of applied CaO-equivalents in dt ha⁻¹, pH recommendations according to VDLUFA (2000a), date of soil sampling, date and depth of tillage in 2017, 2018 and 2019 (in chronological order).

	Struckum	Barlt
Date of lime application	25.08.2016	26.08.2016
Lime1 in CaO dt ha ⁻¹	80	36
Lime1.5 in CaO dt ha ⁻¹	120	54
Start pH	5.0	5.9
Recommended pH level (C)	6.4–7.2	5.9–6.7
Soil Sampling 2017	01.08.	08.08.
Tillage 2017	27.09.	–
Depth [cm]	25	–
Tillage 2018 (1)	25.07./01.08.	27.02.
Depth [cm]	8	30
Soil Sampling 2018	13.11.	14.08.
Tillage 2018 (2)	15.09.	26.09.
Depth [cm]	30	30
Soil Sampling 2019	25.03.	02.04.

5.3.2 Soil sampling and laboratory measurements

Undisturbed soil cores (5 cylinders of 100 cm³, one cylinder of 470 cm³) and disturbed soil samples were taken in each plot, summing up to 24 undisturbed and 4 disturbed samples per treatment and sampling campaign.

The disturbed soil was air-dried and sieved to ≤ 2 mm for chemical analysis. Particle size distribution was derived from a sieve and pipette technique according to DIN ISO 11277 (2002). The pH values were measured in 0.01 M CaCl₂ solution (1:2.5 soil:CaCl₂) according to DIN ISO 11260 (2018).

The exchangeable cations Na⁺, K⁺, Ca²⁺ and Mg²⁺ were extracted in 0.1 M BaCl₂ solution and quantified in an atomic absorption spectrometer (AAS). Additionally, actual acidity was assessed by desorbing H⁺ and Al³⁺ with KCl and determining their concentrations via titration. The effective cation exchange capacity (CEC_{eff}) was determined from the sum of exchangeable cations. The potential cation exchange capacity (CEC_{pot}) was determined with the H-value according to Schachtschabel and

the S-Value (sum of exchangeable cations Na⁺, K⁺, Ca²⁺ and Mg²⁺) (DIN ISO 11260, 2018).

Hydraulic properties

Saturated hydraulic conductivity (k_s) was measured on undisturbed soil cores (100 cm³) based on the falling-head method according to Hartge and Horn (2009).

Unsaturated hydraulic conductivity (k_u) was measured on undisturbed soil cores (470 cm³) with the evapotranspiration-method according to Plagge (1991). Additionally, a k/ψ curve was modeled using the van Genuchten and Mualem model (van Genuchten, 2005) and the software RETC version 6.02 to fit the curve to the measured values solving (eq. 5-1) with the help of the empirical parameters a , n and m .

$$k_u(\psi_m) = k_s \cdot \frac{(1-(a \cdot \psi_m)^{m \cdot n} \cdot (1+(a \cdot \psi_m)^n)^{-m})^2}{(1+(a \cdot \psi_m)^n)^{l \cdot m}} \quad \text{eq. 5-1}$$

with

k_u = unsaturated hydraulic conductivity (cm s⁻¹)

k_s = saturated hydraulic conductivity (cm s⁻¹)

ψ_m = matric potential (kPa)

The pore size distribution was obtained from undisturbed soil cores (100 cm³) by saturating and successively draining them to matric potentials of $\psi_m = -6$, -30 and -50 kPa using ceramic plates and a vacuum device. To derive the fine pore percentage (FP, $\varnothing < 0.2 \mu\text{m}$), small soil cores (2 cm³) were drained to a matric potential of -1500 kPa in an air pressure device (Hartge and Horn, 2009). Bulk density (ρ_B) was determined from the weight to volume ratio after oven-drying at 105°C for 24 h. Total porosity (TP) was determined from ρ_B and solid phase density (eq. 5-2).

$$TP = \left(1 - \left(\frac{\rho_B}{\rho_s}\right)\right) \cdot 100 \quad \text{eq. 5-2}$$

with

TP = total porosity (vol.-%)

ρ_B = bulk density (g cm⁻³)

ρ_s = solid phase density of quartz (2.65 g cm⁻³)

The parameters air capacity (AC, pores $\varnothing > 50 \mu\text{m}$) and plant available water capacity (PAW, pores $\varnothing 50\text{--}0.2 \mu\text{m}$) were derived from the pore size distribution.

Air conductivity

Air conductivity (k_a) was measured on drained soil samples at $\psi_m = -6$ and -30 kPa with an air permeameter under steady-state flow conditions (eq. 5-3) (Peth, 2004).

$$k_a = \frac{\rho \cdot g \cdot q \cdot L_s}{A_s \cdot p} \quad \text{eq. 5-3}$$

with

k_a = air conductivity (cm s^{-1})

ρ = air density (kg m^{-3})

g = gravitational acceleration (m s^{-1})

q = volumetric flow rate ($\text{m}^3 \text{s}^{-1}$)

L_s = length of the sample (m)

A_s = surface of the sample (m^2)

p = pneumatic pressure (Pa)

Air permeability

Afterwards the air permeability (k_{ap}) was calculated from the k_a (eq. 5-4) (Dörner, 2005).

$$k_{ap} = \frac{k_a \eta_a}{\varphi \cdot g} \quad \text{eq. 5-4}$$

with

k_{ap} = air permeability (m^2)

k_a = air conductivity (m s^{-1})

η = air viscosity (Pa s)

ρ = air density (kg m^{-3})

g = gravitational acceleration (m s^{-1})

The defined $\psi_m = -6$ and -30 kPa represent frequently occurring field moisture conditions under which the plant roots are to be supplied with water and air.

Pore continuity index

Pore continuity index C_2 according to Groenevelt et al. (1984) shows the relationship between air permeability (k_{ap}) and air-filled porosity (ε_a) and describes in which extent the air-filled porosity contributes to air permeability (eq. 5–5). High C_2 values indicate a high pore continuity or small tortuosity.

$$C_2 = \frac{k_{ap}}{\varepsilon_a} \quad \text{eq. 5–5}$$

5.3.3 Statistical analysis

The statistical analysis was carried out using the statistical software R version 3.5.1 (R Core Team, 2017). Based on graphical residual analysis, the values of pH, unsaturated hydraulic conductivity (k_u) and the water retention parameters were assumed to be normally distributed and heteroscedastic due to different lime application rates at the site. The values of k_s were not normally distributed. Based on these assumptions, an analysis of variances (One-Factor ANOVA) was determined (Snedecor and Cochran, 1996). All values were evaluated comparing the means of the factor levels treatment and plot for each site separately. The significance of the different tests was set at an α -Level of 5 %. Box-Whisker plots were chosen for the graphical presentation, showing median (mid-line), 25th/75th percentile (lower/upper border of the box), 95th and 5th percentile (Whiskers), 1st and 99th percentile (lower/upper line), outliers (dots) and mean (+).

5.4 Results

5.4.1 Chemical properties

In Struckum, the pH increased with increasing lime application for all limed plots (Table 5-3). The recommended pH-level (C) according to VDLUFA (2000a) was not reached on Lime1 plots, 12-32 months after lime application. The Lime1.5 plots reached the recommended pH-level 24 months after lime application and the pH remained stable even 32 months after lime application (pH-level C according to VDLUFA (2000a)).

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In Barlt, the pH increased with increasing lime application for all limed plots. The recommended pH-level (C) according to VDLUFA (2000a) was reached 12 months after lime application on all limed plots (Lime1 and Lime1.5). The pH values remained almost stable on the limed plots 32 months after lime application.

Table 5-3: pH (CaCl₂ (0.01M), n = 2) for the different lime (CaCO₃) treatments (Lime0 = control, Lime1 = lime recommendation according to VDLUFA, Lime1.5 = lime recommendation according to VDLUFA x 1.5) at both study sites 12, 24 and 32 months after lime application. Values are presented as arithmetic mean.

Site	Treatment	12 months after lime application pH [-]	24 months after lime application pH [-]	32 months after lime application pH [-]
Struckum	Lime0	4.7	5.3	5.4
	Lime1	5.5	6.0	6.2
	Lime1.5	6.3	6.7	6.4
Barlt	Lime0	5.9	5.9	5.8
	Lime1	6.2	6.2	6.0
	Lime1.5	6.3	6.5	6.4

The percentage of exchangeable Ca²⁺ in the topsoil was higher in the limed treatments compared to the control at both study sites (Table 5-4). 32 months after lime application, the percentage of exchangeable Ca²⁺ was still 5–6 % higher in the limed plots compared to Lime0 at the study site in Struckum. Simultaneously, the percentage of exchangeable Mg²⁺ decreased by 4–5 % on the limed plots. The percentage of Mg²⁺ in the exchangeable cation composition is lower at the study site in Barlt compared to the study site in Struckum. The percentage of Ca²⁺ increased by 2 % in the limed plots, but the percentage of Mg²⁺ remained nearly unchanged 32 months after the lime application at the study site in Barlt. The higher soil pH increased the proportion of negative variable charges at the surfaces of e.g. humic substances, and therewith increased the CEC_{eff} in all limed plots, 12 months after the lime application at both sites. The percentage of H⁺ in the limed plots decreased as from 12 months after lime application in Struckum and the decrease was more distinct than at the study site in Barlt. Differences in CEC_{pot} on both study sites can be observed depending on clay content and organic matter.

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Table 5-4: Percentage of exchangeable cations K⁺, Na⁺, Mg²⁺, Ca²⁺, H⁺ and Al³⁺ in the exchangeable cation composition [%]. Effective cation exchange capacity (CEC_{eff}) and potential cation exchange capacity (CEC_{pot}) [cmol_c kg⁻¹] (n = 8) for the different lime treatments (Lime0 = control, Lime1 = lime recommendation according to VDLUFA, Lime1.5 = lime recommendation according to VDLUFA x 1.5) at both study sites 12, 24 and 32 months after lime application. Values are presented as arithmetic mean.

Site	Year	Treatment	K ⁺ %	Na ⁺ %	Mg ²⁺ %	Ca ²⁺ %	H ⁺ %	Al ³⁺ %	CEC _{eff} cmol _c kg ⁻¹	CEC _{pot} cmol _c kg ⁻¹
Struckum	12 months after lime application	Lime0	2.8	1.2	20.4	74.7	0.7	0.1	12.3	-
		Lime1	2.7	1.0	16.3	79.4	0.5	0.2	12.3	21.5
		Lime1.5	2.3	1.1	17.1	79.2	0.3	0.1	14.2	21.6
	24 months after lime application	Lime0	1.7	1.0	20.6	76.0	0.5	0.1	12.0	21.8
		Lime1	1.0	0.9	16.8	80.8	0.4	0.1	12.4	20.3
		Lime1.5	1.6	0.8	15.1	82.3	0.2	0.1	14.1	18.9
	32 months after lime application	Lime0	2.7	1.1	20.5	74.9	0.6	0.2	12.7	20.5
		Lime1	2.4	0.9	15.0	81.5	0.1	0.1	13.6	18.9
		Lime1.5	2.2	0.7	16.7	80.2	0.1	0.0	14.7	22.1
Barlt	12 months after lime application	Lime0	4.0	0.6	9.4	85.3	0.5	0.2	24.3	-
		Lime1	4.0	0.5	9.0	86.2	0.3	0.1	25.2	34.3
		Lime1.5	3.7	0.6	8.9	86.6	0.2	0.1	25.3	33.3
	24 months after lime application	Lime0	4.6	0.7	8.9	85.6	0.2	0.0	23.7	34.2
		Lime1	4.0	0.6	9.1	86.1	0.2	0.0	23.9	34.2
		Lime1.5	4.3	0.5	8.2	86.7	0.2	0.1	24.9	33.4
	32 months after lime application	Lime0	4.3	0.5	9.3	85.7	0.2	0.1	23.9	33.7
		Lime1	4.0	0.4	8.4	86.9	0.2	0.1	25.2	35.0
		Lime1.5	4.2	0.4	9.1	86.1	0.1	0.0	25.4	32.8

5.4.2 Water retention parameters

Twelve months after lime application the average bulk density ρ_B decreased by 9 % in the Lime1.5 treatment relative to the control (Lime0) at the study site in Struckum (Table 5-5). Additionally, plant available water (PAW) significantly increased, and the fine pores (FP) percentage significantly decreased on limed plots (Lime1.5) compared to Lime0 ($p < 0.05$). 24–32 months after lime application, ρ_B , AC, PAW and FP remain almost unchanged in Lime1 and Lime 1.5 treatments compared to the control plots (Lime0).

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At the study site in Barlt, ρ_B decreased by between 2-5 % on all limed plots relative to the control plots 12 to 32 months after lime application. ρ_B remains lower on limed plots compared to the control treatment (Lime0), even 32 months after liming. PAW, FP and AC remain unchanged 12–24 months after lime application on limed plots compared to the control plots (Lime0), while 32 months after lime application, FP significantly decreased on Lime1.5 compared to Lime0 ($p < 0.05$), and AC increased on Lime1 and Lime1.5 compared to Lime0.

Table 5-5: Bulk density (ρ_B , n = 8), plant available water capacity (PAW, n = 8), fine pores (FP, n = 20), air capacity (AC, n = 8) for the different lime treatments (Lime0 = control, Lime1 = lime recommendation according to VDLUFA, Lime1.5 = lime recommendation according to VDLUFA x 1.5) at both study sites 12, 24 and 32 months after lime application. Values are presented as arithmetic mean. Asterisks indicate treatments significantly different from control at $p < 0.05$.

site	treatment	12 months after lime application				24 months after lime application				32 months after lime application			
		ρ_B g cm ⁻³	PAW [vol.-%]	FP	AC	ρ_B g cm ⁻³	PAW [vol.-%]	FP	AC	ρ_B g cm ⁻³	PAW [vol.-%]	FP	AC
Struckum	Lime0	1.40	5	34	9	1.32	7	29	13	1.25	14	25	13
	Lime1	1.38	8	29*	10	1.32	8	28	14	1.23	12	26	15
	Lime1.5	1.26	14*	29*	9	1.31	7	30	14	1.29	14	26	11
Barlt	Lime0	1.15	14	31	11	1.19	5	37	13	1.12	15	34	9
	Lime1	1.15	14	31	10	1.14	5	38	14	1.07	16	32	12
	Lime1.5	1.15	12	32	13	1.17	5	38	13	1.09	16	30*	13

5.4.3 Water and air transport parameters

Hydraulic conductivity

Twelve months after lime application, there is little difference between the saturated hydraulic conductivity (k_s) in the limed plots and the control plots (Lime0) at the study sites in Struckum and Barlt (Fig. 5-2). However, k_s clearly increased 24-32 months after lime application on both limed plots compared to Lime0 in Struckum, and significantly increased 24 months after lime application on Lime1 compared to Lime0 ($p < 0.05$) in Barlt.

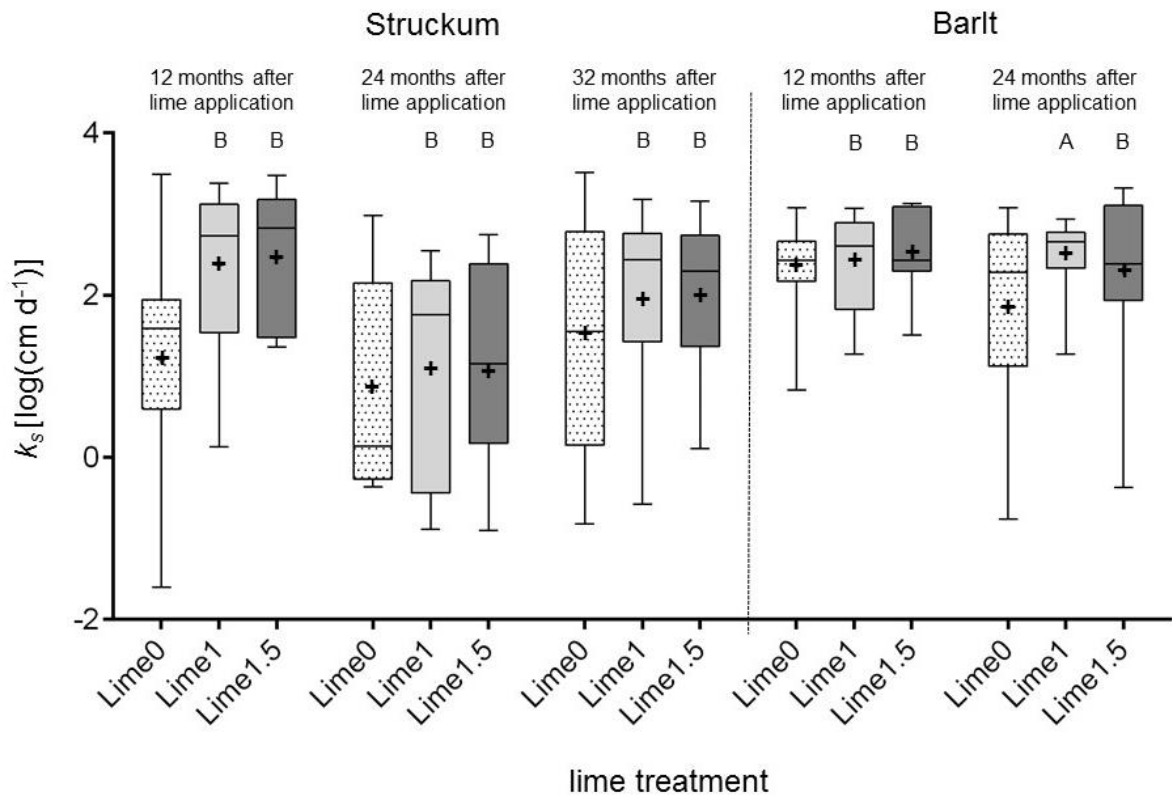


Fig. 5-2: Saturated hydraulic conductivity k_s [cm d^{-1}] (logarithmic display) for the different lime treatments (Lime0 = control, Lime1 = lime recommendation according to VDLUFA, Lime1.5 = lime recommendation according to VDLUFA x 1.5) at both study sites 12, 24 months after lime application and for Struckum 32 months after lime application. Values are presented as geometric mean. Crosses denote arithmetic means. $n = 12$, A (B) indicates treatment (not) significantly different from the control (Lime0) at $p < 0.05$.

Fig. 5-3 provides a more detailed view on the hydraulic conductivity (k) dynamics for the study site in Barlt 32 months after lime application. Under water saturated conditions ($\psi_m = 0$ kPa), k is higher on Lime1 compared to the control plots (Lime0) while Lime1.5 is equal to Lime0. For $\psi_m = -6$ kPa, the highest k was found in the Lime1 plots compared to Lime0. Similarly, a higher water transport under unsaturated conditions was measured at the Lime1.5 plots for $\psi_m = -6$ kPa compared to Lime0. However, the differences among the three lime treatments were not significant and for $\psi_m = -30$ kPa, the lime treatments Lime1 and Lime1.5 are showing no differences compared to Lime0.

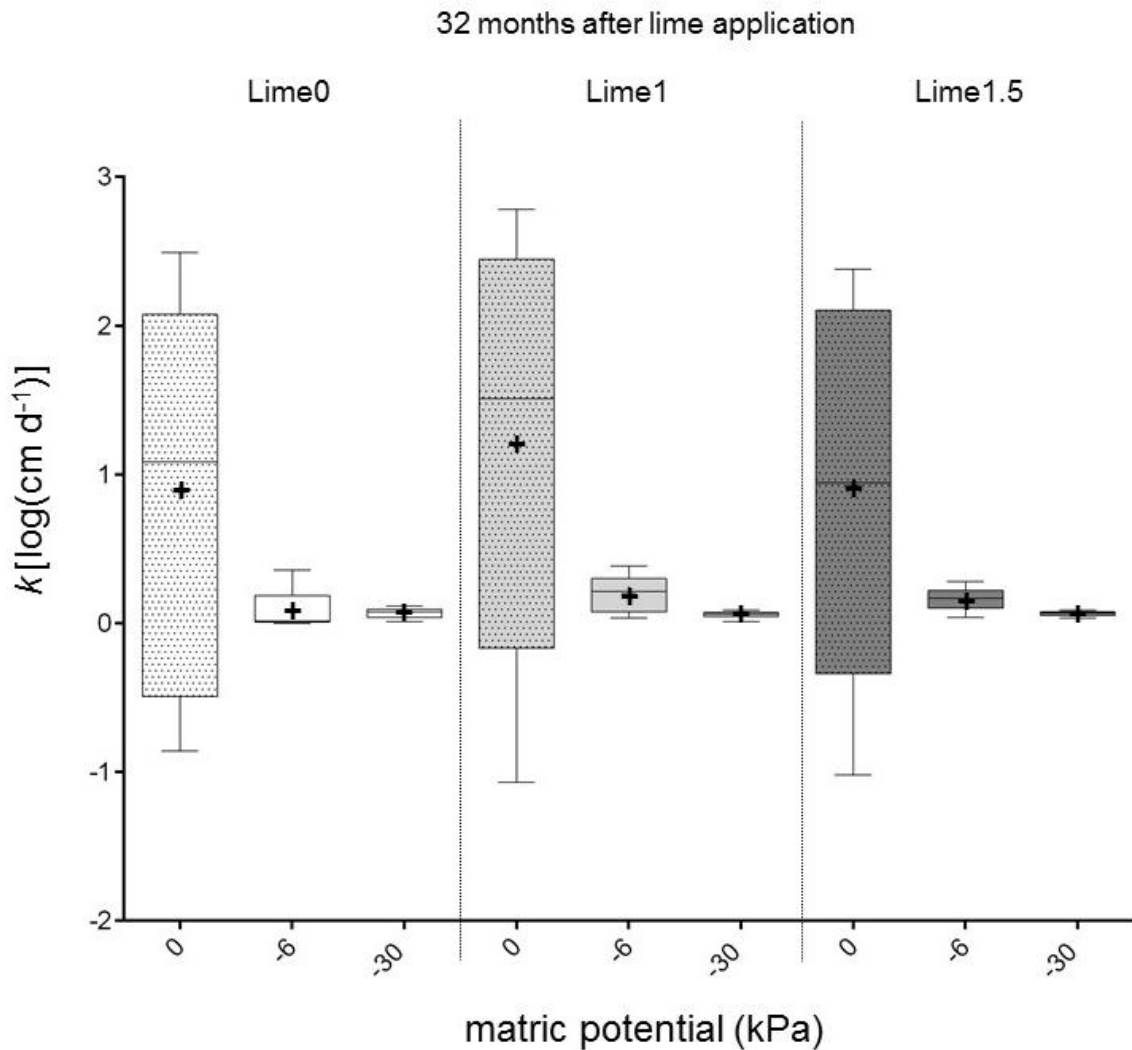


Fig. 5-3: Hydraulic conductivity k [cm d^{-1}] (logarithmic display) at the matric potentials 0 kPa, -6 kPa and -30 kPa for the different lime treatments (Lime0 = zero lime, Lime1 = lime recommendation according to VDLUFA, Lime1.5 = lime recommendation according to VDLUFA x 1.5) at the study site in Barlt 32 months after lime application. Crosses denote arithmetic means. $n = 12$ (0 kPa), $n = 12$ (Lime0, -60 kPa), 11 (Lime0, -30 kPa), 17 (Lime1, -60 kPa), 18 (Lime1, -30 kPa), 21 (Lime1.5, -60 kPa), 12 (Lime1.5, -30 kPa).

Air-filled porosity

ε_a did not change significantly in the limed treatments (Lime1 and Lime1.5) in the topsoil at both matric potentials compared to the control plots (Lime0) at the study site in Barlt (Fig. 5-4). However, a slight increase is visible in the limed treatments compared to the control plots (Lime0).

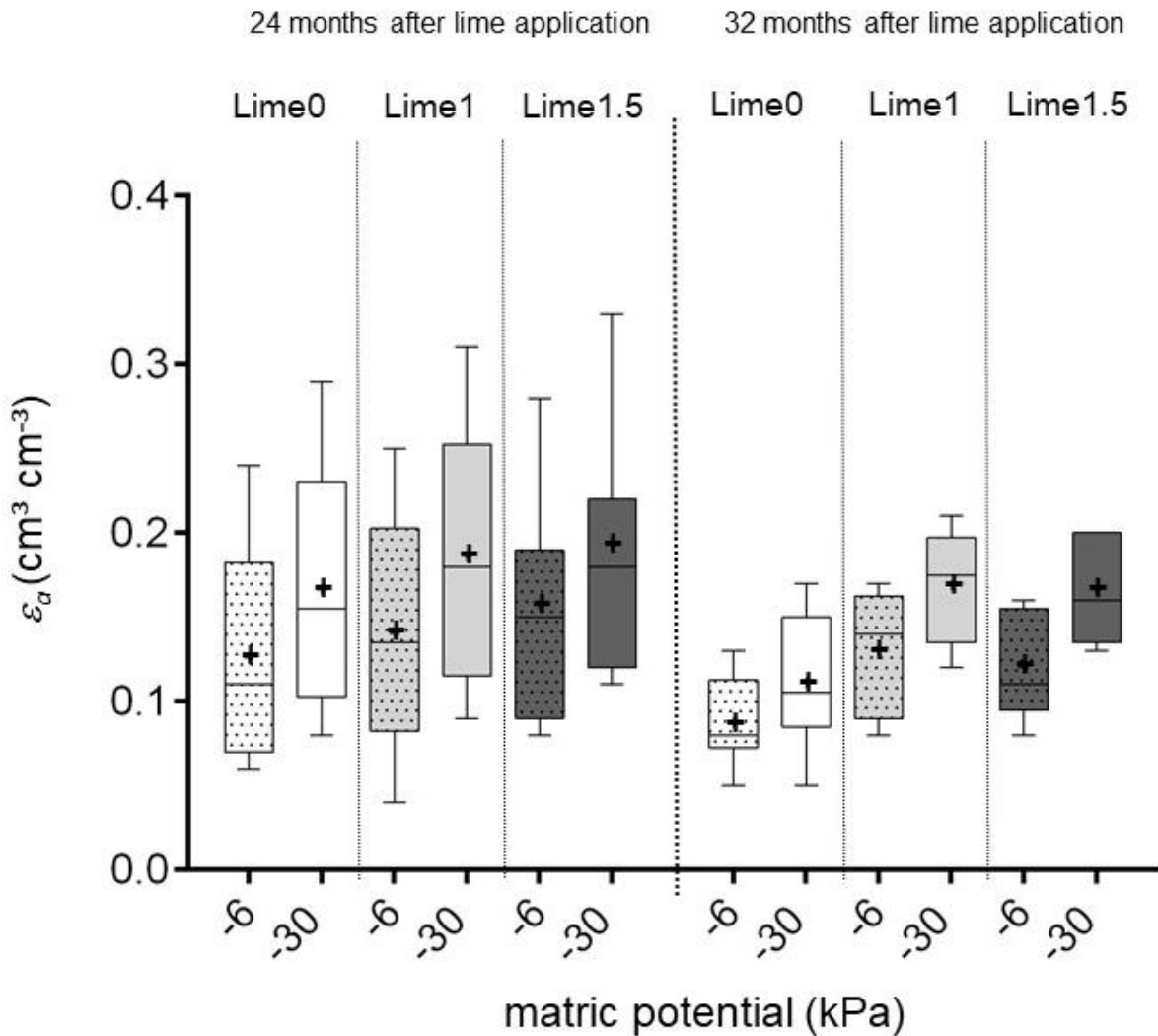


Fig. 5-4: Air-filled porosity ε_a in $\text{cm}^3 \text{cm}^{-3}$ at matric potentials of -6 kPa (corresponding to AC, Table 5-5) and -30 kPa for the different lime treatments (Lime0 = zero lime, Lime1 = lime recommendation according to VDLUFA, Lime1.5 = lime recommendation according to VDLUFA x 1.5) at the study site in Barlt 24 months and 32 months after lime application. Crosses denote arithmetic means. (n = 8)

Air permeability

Air permeability (k_{ap}) did not change significantly in the limed plots (Lime1 and Lime1.5) compared to the control (Lime0) 24 and 32 months after lime application at a matric potential of -30 kPa at the study site in Barlt (Fig. 5-5). However, a slight increase is visible in the limed treatments compared to the control plots (Lime0).

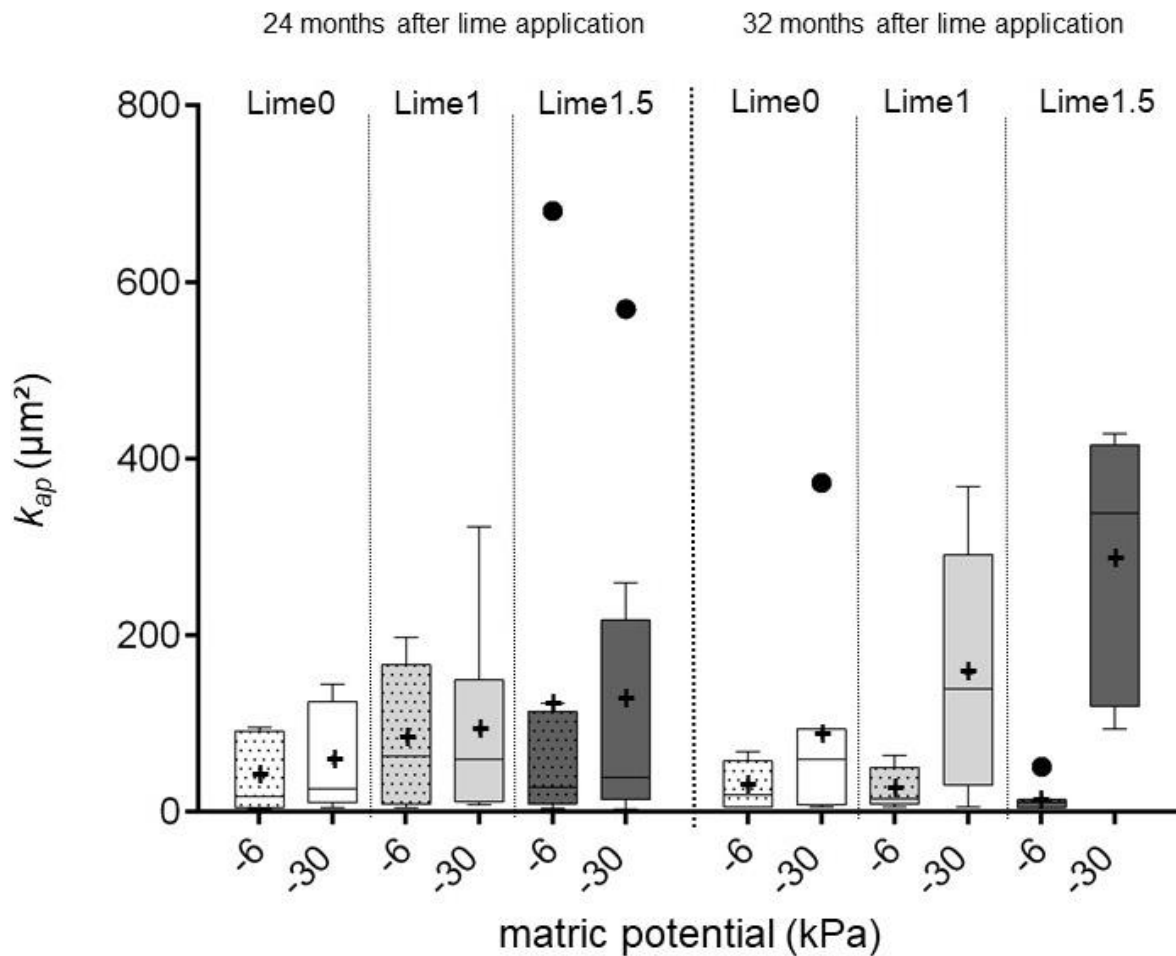


Fig. 5-5: Air permeability k_{ap} in μm^2 for the different lime treatments (Lime0 = zero lime, Lime1 = lime recommendation according to VDLUFA, Lime1.5 = lime recommendation according to VDLUFA x 1.5) at the study site in Barlt, 24 months and 32 months after lime application. Crosses denote arithmetic means. (n = 8)

Pore continuity index

Pore continuity, represented by the index C_2 , increased in the limed plots (Lime1 and Lime1.5) compared to Lime0 at -6 kPa and -30 kPa 24 months after the lime application, and at -30 kPa 32 months after lime application (Fig. 5-6). The highest C_2 values were determined in the Lime1.5 treatment.

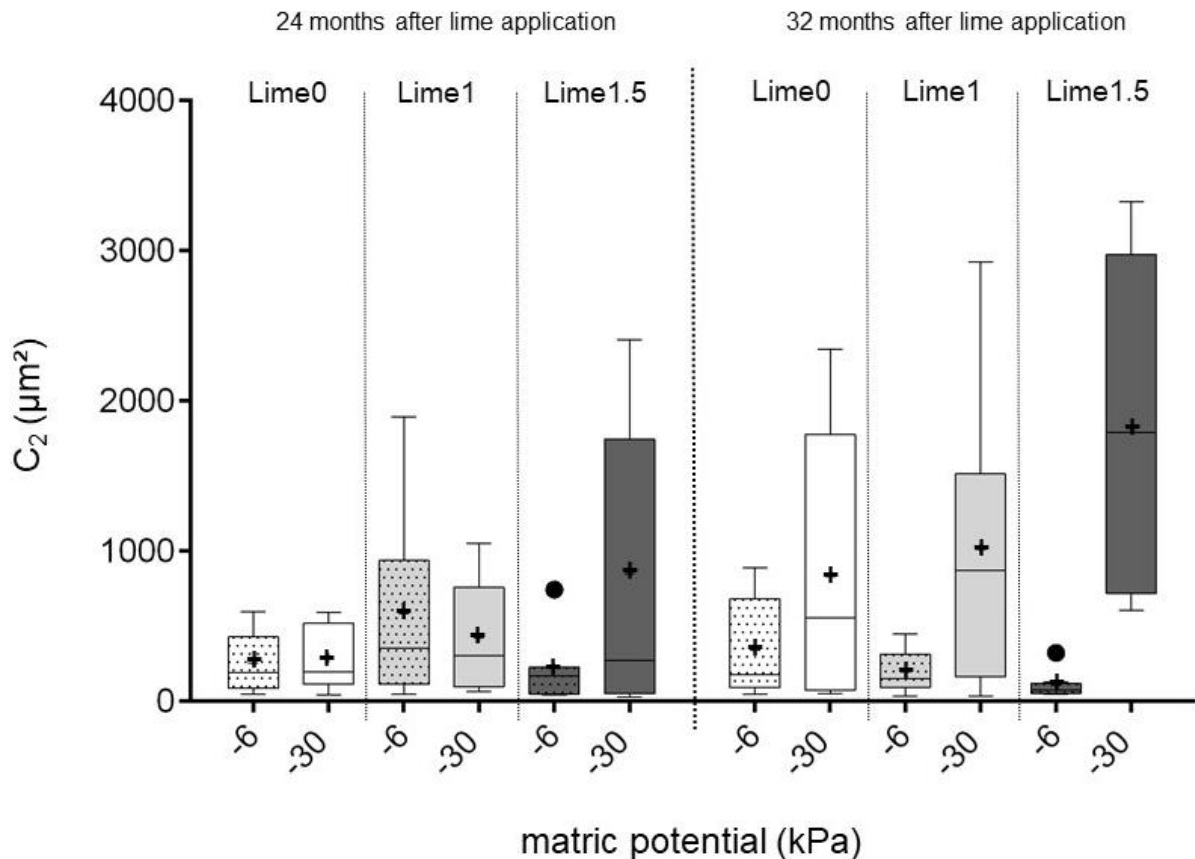


Fig. 5-6: C_2 (pore continuity) indices in μm^2 at matric potentials of -6 and -30 kPa for the different lime treatments (Lime0 = zero lime, Lime1 = lime recommendation according to VDLUFA, Lime1.5 = lime recommendation according to VDLUFA x 1.5) at the study site in Barlt, 24 months and 32 months after lime application. Crosses denote arithmetic means. (n = 8)

5.5 Discussion

Lime application on arable land is a worldwide practice to ameliorate acidic and/or structurally damaged soils. The major areas of application are in the tropics and sub-tropics because of the intensive weathering processes in these soils (Auler et al., 2017a). It is well known that the acidification process reduces plant growth, water and nutrient uptake and yields because of a low content of base cations, especially calcium (Caires et al., 2008), and the sensitivity of soils to inadequate nutrient status and soil

strength cause problems in arable soils worldwide. Especially in the relatively young and often fine-textured marshland soils in northern Germany, the functionality of the actual pore system, both with respect to physicochemical and physical processes, is increasingly endangered, because the often high clay content and long periods with water saturation throughout the year render the soil very susceptible to compaction, and a low aggregate stability supports surface sealing (Burbaum, 2008). Thus, on these sites, soil structure needs to be ameliorated and maintained at an optimal level to obtain beneficial conditions for root growth and provide high crop yields. While farmers fertilize their sites regularly, the application of limestone is often minimal. This can be explained by a widespread underestimation of the enormous effects that a functioning pore system and optimized water and air fluxes in well-structured soils have on crop yield, and by a lack of knowledge on the potential of lime applications to ameliorate soil structure functionality.

The elevation of the pH-value was successful on all limed plots subsequent to the lime application on both survey sites. In Barlt, the target pH-value according to VDLUFA (2000a) was reached on the limed plots 12 months after lime application, and remained stable during the surveyed three years after lime application. For Struckum, the amount of applied lime was not high enough to reach the recommended pH-range according VDLUFA (2000a), because of restrictions on the maximal annual amounts of lime application in Schleswig-Holstein. Although the amount of applied lime the recommendations according to VDLUFA (2000a) in the Lime 1.5 treatment – it was not possible to reach the recommended pH-range in this treatment either, due to a reduced accessibility of (clay) particle surfaces for Ca^{2+} . Nevertheless, the lime applications successfully increased soil pH, CEC_{eff} , PAW, and soil aeration parameters, and lastingly decreased ρ_B on both sites.

The elevated pH increased the negative variable particle charge and thus CEC_{eff} . The components of the dissolved lime increase ionic strength in the soil solution, and the resulting compression of the diffuse double layers around soil colloids as well as cation bridging lead to flocculation and aggregation (Oades, 1984b; Voelkner et al., 2015). Ion exchange plays a vital role in the storage of plant-available nutrients in soils. Owing to the predominantly negative charge in temperate region soils, especially cations play

an important role in affecting soil particle interaction, and, with that, soil structure (Holthusen, 2010; Wuddivira and Camps-Roach, 2007). Lime applications can lastingly increase the concentration of exchangeable Ca^{2+} in the soil solution (Valzano et al., 2001). In our results, the percentage of Ca^{2+} in the exchangeable cation composition increased with increasing amount of applied lime 12 months after application and remained elevated throughout the three years of the survey. Liming of acid soils enhances the substitution of exchangeable Al^{3+} by Ca^{2+} and thereby reduces aluminum toxicity (Caires et al., 2008). Correspondingly, the lime application realized a permanent reduction of the acid cations H^+ and Al^{3+} in the exchangeable cation composition at both study sites. The clay mineral montmorillonite has a preference for Ca^{2+} over Mg^{2+} while illite has a preference for Mg^{2+} over Ca^{2+} in the adsorption complex (Rengasamy et al., 2016). Accordingly, the soil at the study site in Struckum, with the dominate clay mineral illite, exhibits a higher amount of exchangeable Mg^{2+} compared to the study site in Barlt with both illite and montmorillonite clay minerals. However, the increased percentage of Ca^{2+} in the soil solution after the lime application reduced the percentage of Mg^{2+} in the cationic exchange complex, especially on the limed plots at the study site in Struckum. Exchangeable Mg^{2+} increases the extent of the diffuse double layer around soil colloids more than exchangeable Ca^{2+} (Curtin et al., 1994; Keren, 1991) and therefore, high amounts of exchangeable Mg^{2+} can increase clay dispersion and reduce soil structural stability e.g. by increasing the vulnerability of soil aggregates to raindrop impact. The consequence of such dispersion is smaller aggregates, which will block pores and promote further surface sealing during rainfall. This is in agreement with the findings of Dontsova and Norton (2001), who documented that surface sealing is less distinct in Ca-saturated than in Mg-saturated soils. Our results also prove that high amounts of fine-grained limestone (CaCO_3) can change the Ca^{2+} and Mg^{2+} ratio under humid climate conditions and thus, reduce the danger of dispersion in the clayey soils with related risk of erosion.

Large hydrate shells of Mg^{2+} cause clay swelling, and swelling in turn reduces pore diameters and promotes aggregate disintegration. Consequently, lower k_s occurs (Markgraf et al., 2012; Zhang and Norton, 2002). Our results agree with these findings, as increasing k_s is detected with increasing lime dosage and can be linked with the

increasing replacement of Mg^{2+} by Ca^{2+} in the exchangeable cation composition 12 months after lime application in Struckum.

The question concerning the optimal amount of lime application considering the physical strengthening and improved hydraulic functions requires some further discussion. It is well known that liming can improve PAW and AC and reduce the percentage of FP in marshland soils (Bartels et al., 1985). Accordingly, we discovered significantly lower percentages of FP and higher PAW at the study site in Struckum, already one year after the amelioration with lime. However, the changes were non-persistent, as 24 months after lime application PAW, FP and AC were on the same level in the control and the lime treatments. On this site, the lime amount corresponding to lime recommendation according to VDLUFA (2000a) x 1.5 exceeded the permitted annual lime application amount for Schleswig-Holstein. Consequently, the Lime1.5 treatment on this site is equivalent to the lime recommendation according to VDLUFA (2000a), while the Lime1 treatment corresponds not to lime recommendation according to VDLUFA (2000a). Apparently, given the low start pH in Struckum, even the amount of applied lime corresponding with the lime recommendation according to VDLUFA (2000a) (Lime1.5 treatment) was not high enough to change the water retention parameters sustainably. At the study site in Barlt the application of lime amounts corresponding to the lime recommendation according to VDLUFA (2000a) x 1.5 was possible, and we found a significantly lower percentage of FP and a higher AC throughout the survey time of 32 months in both lime treatments. Obviously, on this site, the amount of limestone that corresponded with the recommendations of VDLUFA (2000a) was sufficient to change the parameters persistently, and an increase of the lime amount corresponding to VDLUFA (2000a) recommendation x 1.5 would not have been necessary for changing some physical parameters.

Surface application of lime on a silty-clayey topsoil can increase TP and reduce ρ_B within 18 months without tillage (Auler et al., 2017). Our results indicate that an increase in TP and a reduction of ρ_B also occurs if the lime is incorporated by tillage. Additionally, the lime applications shifted the pore size distribution from finer pores towards narrow coarse pores. Further, lime applications lead to a higher aggregate stability and thereby promotes the development of a stronger structure (Aye et al.,

2016), which helps to avoid homogenization and the formation of large clods during ploughing. The improved aggregation and increased porosity following the lime application (Kirkham et al., 2007) enhances the k_s and thus, the availability of water (Horn and Smucker, 2005). In our experiment, both the k_s and the k_u was improved by the application of lime (Lime1 and Lime1.5) at the study site in Barlt. This represents an improved availability and subsequent delivery of water for plants during dryer times. However, our results also show that tillage effects intensely overlap the strengthening effects as derived from the changes of the hydraulic conductivity with time. The strong reduction of k_s in all treatments 24 months after lime application proves that the tillage event four months before the sampling at the site in Struckum destroyed the pore continuity (hydraulic conductivity) intensely. Even the improved pore connectivity in the limed plots was not rigid enough to withstand the tillage effects. However, the decline was less intense than in the control, suggesting a higher rigidity of the soil structure towards mechanical disturbance. Soil aggregates are vulnerable to disruption by physical disturbance such as tillage (Bronick and Lal, 2005), and therefore tillage directly affects porosity and pore continuity (Dörner et al., 2012). On the other hand, reduced tillage can increase macro-porosity and thereby enhance water and air movement in soil (Ehlers, 1977; Warkentin, 2001; Wiermann et al., 2000). Lime incorporation by tillage can help to obtain an optimal effect concerning the distribution of the lime within the soil and the interactions of the lime components with particle surfaces. However, the time demanding strengthening of particle bonds requires the avoidance of tillage impacts. Thus, the immediate incorporation of the lime directly after application is advisable (Caires et al., 2008; Joris et al., 2016), while the following repeated disturbance of the developing soil structure is considered as harmful. Furthermore, we need to consider the regionally different amounts of seepage water, because the effects of surface-applied lime are often limited to the upper centimeters of the topsoil and deeper horizons react retarded depending on the transport of Ca^{2+} ions towards the subsoil (Joris et al., 2016).

Consequences for the ρ_B are more obvious for Barlt compared to Struckum 24 months after lime application, because of the higher start-pH and applied amount of lime. ρ_B is often inversely related to TP and partly to hydraulic conductivity, which implies that a

reduction in ρ_B causes an increase in the TP and may increase the hydraulic conductivity of the soil. k_{ap} and k_s are useful parameters for the evaluation of these strengthening processes of the soil structure (Reszkowska et al., 2011), because the elevation of these parameters is related to the new structure formation (Mordhorst et al., 2018).

In structured soils compared to homogenized soils, k_{ap} and k are significantly higher because of the major role of the macropores in the soil system (Tuli et al., 2005). Transport of air however depends not only on the actually available ε_a , but also on the continuity of these pores as a function of soil structure. The application of lime increases the k_a and the ε_a at a given ψ_m , presumably because the flocculation of clay particles and the formation of bridges between them (Oades, 1984b) enhances the formation of aggregates with a well-connected inter-aggregate pore system. The C_2 index describes how increases in ε_a contribute to k_{ap} , and therefore delivers information on pore continuity (Groenevelt et al., 1984). The more distinct increase in the C_2 indices from $\psi_m = -6$ to $\psi_m = -30$ kPa in the limed plots compared to the control 32 months after lime application indicates an improved pore continuity, because the addition of the narrow coarse pores to ε_a improved k_{ap} more in the limed plots than in the control. The determined results on the k_a for the limed plots had similar trends as k_s , because of the major role of soil structure and the development of macro-pores in air and water transport processes in the soil.

5.6 Conclusions

Lime applications to marshland soils generated a well-connected pore system with increased k_s due to a shift in the pore size distribution from finer pores towards coarser pores and an overall increase in porosity.

Liming led to the substitution of other exchangeable cations by Ca^{2+} on the surfaces of the soil colloids, and thereby reduced the swelling intensity of the clay minerals. In marshland soils with a high clay content, the amount of applied lime according to the current recommendations - depending always on the initial pH - was not enough to induce a long-term change in the soil structure.

Especially for fields with a low start pH-value and high clay content, a higher amount of applied lime and repeated applications (for example two applications in two consecutive years) are necessary. Another method could be to apply 50 % of the dosage before the first management operation (tillage like plowing or harrowing) at the beginning of the season and the remaining 50 % after this operation.

The current lime recommendations must be improved by considering additional soil physical and chemical factors such as PAW and CEC_{eff} . Without tillage, a recovery of the porosity and the creation of a stable pore network will more likely occur, and consequently, this should be the preferred procedure.

We recommend the incorporation of lime by ploughing right after application, but only reduced tillage in the following years to allow for undisturbed soil structure development.

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6 Effect of CaCO₃ application on mechanical strength and hydraulic properties in arable soils

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submitted to Soil Use and Management

6.1 Abstract

Liming is a well-established management procedure to enhance the productivity of acidic arable soils. However, approx. 42 % of the arable mineral soils in Germany are undersupplied with lime. Especially in clay-rich soils, lime applications will not only increase soil pH but also ameliorate soil structure. Therefore, the influence of lime application on soil structure was examined in three field trials with various soil texture and initial pH values. Each field trial included three lime treatments: a control with no lime application (Lime0), lime application according to the soil-specific recommendations by VDLUFA (2000a) to achieve an optimal soil pH for plant production (Lime1), and VDLUFA (2000) recommendations x 1.5 (Lime1.5). The lime application rates were equivalent of 25 dt CaO ha⁻¹ (Neubrandenburg), 36 dt CaO ha⁻¹ (Bartl) and 98 dt CaO ha⁻¹ (Schrecksbach) in the “Lime1”-treatment and 37.5 dt CaO ha⁻¹ (Neubrandenburg), 54 dt CaO ha⁻¹ (Bartl) and 147 dt CaO ha⁻¹ (Schrecksbach) in the “Lime1.5”-treatment. We hypothesize that hydraulic and air conductivity, pore size distribution, shear parameters (cohesion and angle of internal friction) as well as aggregate tensile strength are useful indicators to quantify changes in soil structure initiated by lime applications.

The results show favorable changes in soil structure, which enhance water and air flow in the limed plots. Moreover, cohesion and shear strength of the soil matrix increase a few months after lime application at all study sites. Additionally, an increasing tensile strength of air-dried and pre-drained aggregates is observed. However, due to the annual tillage, a major proportion of the strengthening effects is destroyed. This results in following recommendations: (i) soil tillage should be avoided in order to support the continuous structure strengthening, and (ii) repeated lime applications should be

implemented successive years in order to achieve a permanent and sustainable structural effect in soils.

6.2 Introduction

The improvement of soil management practice is essential for the sustainable use of arable land worldwide. To achieve this, soil properties must be carefully considered, and future management strategies must be adapted accordingly. Soil structure and its rigidity play an important role in the management of soil resources and should be included in the process of developing a sustainable agriculture (Horn et al., 2014).

Soil structure is defined by the spatial arrangement of particles and pores (Díaz-Zorita et al., 2002; Oades, 1984b), and results from the rearrangement of particles due to capillary forces, as well as from the flocculation, bonding and cementation of mineral particles with inorganic and organic substances (Six et al., 2000; Tisdall and Oades, 1982). Aggregate formation thus depends on changes in soil water content and the coinciding varying hydraulic potential, defined as the sum of matric and gravitational potential, due to wetting and drying during the year, as well as chemical processes and biological activity in the soil. Capillary forces draw particles towards each other and thereby increase the number of contact points (Dexter and Horn, 1988). The particle rearrangement initially increases the aggregate bulk density and the strength of the individual aggregates, which leads towards an optimal particle organization within single aggregates and eventually results in smaller aggregate bulk densities combined with higher soil strength (Baumgartl and Horn, 1991; Horn and Smucker, 2005). The stability, size distribution and spatial organization of soil aggregates influence pore geometry (Lipiec et al., 2009) and the overall stability of the soil structure (Kay, 1990).

The alterations of soil structural stability due to mechanical impacts, as well as the naturally occurring continuous fluctuations throughout the year are well documented (Hartge and Horn, 2009; Horn, 2004; Horn and Lebert, 1994). Soil shear strength quantifies the resistance towards shear deformation under mechanical impact, defines the internal soil strength directly, and can be assessed from the shear parameters: angle of internal friction and cohesion. While both values increase during the progressing, undisturbed structure formation, homogenization and kneading can lead

to shear failure, which is furthermore supported by pore water menisci shapes changing towards convex and results in a decline of cohesion and angle of internal friction. Shear failure leads to the complete blockage of formerly connected transport paths, and thereby inhibits water and gas fluxes (Horn et al., 2003). Soil characteristics, such as hydraulic and air conductivity, air capacity (percentage of pores with $\varnothing > 50 \mu\text{m}$) and plant available water capacity (percentage of pores with $\varnothing 0.2\text{--}10 \mu\text{m}$) are often negatively affected by mechanical impact, e.g. by traffic on arable land, because soil compaction results in changes of the pore size distribution with a reduction of wide and narrow coarse pores (Gebhardt et al., 2009) and a reduced pore continuity (Dörner, 2005; Reszkowska, 2010).

Soil aggregates persist as long as their strength is higher than the given load or shrinkage forces (Horn and Peth, 2009). Otherwise, soil fragmentation, the breaking of aggregates into finer units, occurs (Díaz-Zorita et al., 2002). Apart from natural processes such as shrinkage and swelling, the disintegration of aggregates into smaller units can be caused by plowing and excavations in the field (Horn and Smucker, 2005). The destruction of soil aggregates can result in reduced water infiltration, air fluxes, nutrient buffering functions and downgraded soil fertility (Horn and Peth, 2012), as well as enhanced mineralization of previously physically protected organic matter, followed by e.g. increasing emissions of greenhouse gases (Caires et al., 2006; Sheehy et al., 2015; Six et al., 2000). On the other hand, surface liming in a no-till system, without disruption of the pore system, can promote soil aggregation and increase the accumulation of organic matter in different aggregate size classes (Bossolani et al., 2021; Hati et al., 2008). This physical protection of organic matter or nutrients in aggregates (Briedis et al., 2012b) can reduce greenhouse gas emissions and thus be an important factor to mitigate climate change. Consequently, a high aggregate stability is desirable, especially for arable soils that are subjected to tillage. Soil properties, such as organic carbon content, ion content and composition in the soil solution, texture, bulk density, the intensity and number of swelling and shrinkage cycles, particle mobility and bonds between particles in or between the aggregates influence aggregate strength (Dexter, 1988; Park and Smucker, 2005a; Semmel et al., 1990). Cementing agents (e.g. calcite) especially affect the strengthening and the bonding between mineral particles, and organic matter further enhances the resistance

against soil deformation (Bronick and Lal, 2005). Finally, also several biological binding agents, such as fungal hyphae and microbial exudates, strengthen soil aggregates and in conclusion also soil structure and its rigidity (Abiven et al., 2009; Tisdall and Oades, 1982). A sensitive indicator for soil physical quality is the tensile strength of aggregates (Dexter and Kroesbergen, 1985), because it reflects the resistance against soil deformation in the weakest failure zones (Braunack et al., 1979).

It is well known that lime application neutralizes the acidity in soils and helps to increase aggregate stability, which in turn leads to an improved soil structure (Bell, 1996; Goulding, 2016; Hartge and Bohne, 1985; Haynes and Naidu, 1998; Metzger et al., 2020). However, Lausen (2012) and Stolpe and Schimpf (2017) emphasize an urgent need of increased lime application in different federal states in Germany, because e.g. 50 % of the clayey arable soils in Schleswig-Holstein as well as > 25 % of the arable soils in Saxony-Anhalt are still undersupplied. Jacobs et al. (2018) reported that approx. 42 % of the mineral arable soils and 57 % of the soils under permanent grassland in Germany are not sufficiently supplied with lime. In Germany, recommendations for lime application rates consider the deviation of the current soil pH (CaCl₂) from a target pH value that is considered optimal for the respective soil texture and organic matter content (VDLUFA, 2000a). According to VDLUFA (2000), optimal conditions for soil structure and nutrient availability are given at the Level "C", but this concept does not consider the potential benefits for soil physical properties and functions mentioned above. When limestone (CaCO₃) is applied to soil, it reacts with protons (H⁺) in the soil solution and the Ca²⁺-ion is released, while the protons are bound in the formation of hydrogen carbonate (eq. 6-1).



The released Ca²⁺ can promote the exchange and leaching of acidic cations (H⁺, Al³⁺) from negatively charged surfaces (Beetham, 2015). Additionally, the released Ca²⁺ forms clay-cation bonds, which can be completely covalent or ionic or a combination of both (Rengasamy et al., 2016). The increase of the ionic force along with the greater availability of soluble Ca²⁺ in the soil promotes a thinning of the diffuse double layer around soil colloids (Sparks, 2003), representing the fractions (< 2 µm) of the soil including clay and organic matter with a large surface area (Gavrilescu, 2014). When

the diffuse double layer surrounding the clay colloids is compressed, clay particles are more likely to join into flocs, enter face-to-face associations and form clusters (Dash and Hussain, 2015), or connect with negatively charged organic colloids with the assistance of Ca²⁺ (Beetham, 2015; Haas et al., 2018b). The formation of stable aggregates, which is supported by the flocculation and agglomeration of clay particles (Beetham, 2015), enhances physical properties such as water holding capacity (Hoyt, 1981) and creates a finer tilth in the seedbed of arable soils (Blomquist et al., 2017). Especially clay soils benefit greatly from a structural improvement through lime application. Soils with a dominance of expansive montmorillonite clay minerals furthermore exhibit a high cation exchange capacity and are expected to respond more rapidly to lime treatment than those with kaolinite as the dominant clay mineral (Bell, 1996). Liming also reduces the tendency to silting and erosion, and the soil becomes more stable and less susceptible to compaction, which at the same time supports plant root growth (Castro, 1991).

In general, field trial research that considers the influence of lime application on soil physical properties and aggregate stability is rare, especially in recent years in Germany. Therefore, the effects of lime applications on soil structure functionality and soil strength, with special focus on aggregate stability, in arable soils are highlighted in this work.

The following key questions were determined for our study:

- Can an increased lime application rate alter the shear parameters in agricultural soils, and how is the shear strength affected by texture, soil organic matter and clay mineral composition?
- To which extent do changes in soil pore size distribution remain detectable 12 and 24 months after lime application with tillage between the two sampling times?
- Is there an influence of lime application on tensile strength of air-dried and pre-drained aggregates?

6.3 Material and methods

6.3.1 Experimental design and soil sampling

Three field trials on soils with very different texture and structure were established on arable land in summer 2016 and spring 2017. The experimental design included three treatments: a control with no lime application (Lime0), lime application according to the soil-specific recommendations by VDLUFA (2000a) to achieve an optimized soil pH (Lime1), and soil-specific recommendations x 1.5 (Lime1.5). The applied fine-grained limestone contains low amounts of magnesium, and the neutralizing value is 48 %. The treatments were arranged in a randomized block design with four replicates. Every plot had a size of 6 m x 18 m, and local farmers managed the fields conventionally with annual tillage and nitrogen mineral fertilizers.

The first field trial “Neubrandenburg” is in the north-eastern part of Germany in the state Mecklenburg-Western Pomerania. The soil type is a Haplic Luvisol according to the IUSS Working Group WRB (2014). The initial pH (in CaCl₂, before lime application) is 5.8, and the recommended pH is between 6.3 and 7.0 (VDLUFA, 2000a). The lime application rates were equivalent to 25 dt CaO ha⁻¹ in the “Lime1”-treatment and 37.5 dt CaO ha⁻¹ in the “Lime1.5”-treatment. The lime was applied and incorporated with a disc harrow (~ 8 cm) only once at the beginning of the field trial. The crops were maize (2018) and winter barley (2019). Soil sampling took place 12 (2018) and 24 (2019) months after lime application from the topsoil (0–30 cm depth) of the Lime0, Lime1 and Lime1.5 plots and with tillage between the two sampling times (Table 6-1).

The second field trial “Barlt” is in the north of Germany in the state Schleswig-Holstein. The soil is a Haplic Gleysol according to the IUSS Working Group WRB (2014). The initial pH is 5.9, and the recommended pH is between 5.9 and 6.7 (VDLUFA, 2000a). The lime application rates were equivalent of 36 dt CaO ha⁻¹ in the “Lime1”-treatment and 54 dt CaO ha⁻¹ in the “Lime1.5”-treatment. The lime was applied and incorporated with a plough (~ 25 cm) only once at the beginning of the field trial. The crops were winter wheat (2017) and summer wheat (2018). Soil sampling took place 12 (2017) and 24 (2018) months after lime application from the topsoil (0–30 cm depth) of the

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Lime0, Lime1 and Lime1.5 plots and with tillage between the two sampling times (Table 6-1).

The third field trial “Schrecksbach” is in central Germany in the state of Hesse. The soil is a Haplic Stagnosol according to the IUSS Working Group WRB (2014). The initial pH is 5.3, and the recommended pH ranges between 6.4 and 7.2 (VDLUFA, 2000a). The lime application rates were equivalent of 98 dt CaO ha⁻¹ in the “Lime1”-treatment and 147 dt CaO ha⁻¹ in the “Lime1.5”-treatment. The lime was applied and incorporated with a disc harrow (~ 8 cm) only once at the beginning of the field trial. The crops were winter wheat (2018) and winter wheat (2019). Soil sampling took place 12 (2018) and 24 (2019) months after lime application from the topsoil (0–30 cm depth) of the Lime0, Lime1 and Lime1.5 plots and with tillage between the two sampling times (Table 6-1).

Table 6-1: Date of lime application, quantity of applied CaO-equivalents in dt ha⁻¹, pH recommendations according to VDLUFA (2000), date of soil sampling, date and depth of tillage in 2017-2019 (in chronological order).

	Neubrandenburg	Barlt	Schrecksbach
Date of lime application	03.04.2017	26.08.2016	06.04.2017
Lime1 in CaO dt ha ⁻¹	25	36	98
Lime1.5 in CaO dt ha ⁻¹	37.5	54	147
Initial pH	5.8	5.9	5.3
Recommended pH level (C)	6.3–7.0	5.9–6.7	6.4–7.2
Tillage 2017 (1)	03.05.	–	04.04.
Depth [cm]	28	–	8
Soil Sampling 2017	–	08.08.	–
Tillage 2017 (2)	16.10.	–	09.10.
Depth [cm]	25	–	25
Soil Sampling 2018	10.03.	14.08.	19.04.
Tillage 2018	15.10.	27.02.	08.10.
Depth [cm]	22	30	25
Soil Sampling 2019	14.04.	–	09.04.

6.3.2 Measurements on homogenized soil samples

For all three study sites, the following soil characteristics were analyzed on homogenized, air-dried soil material sieved to ≤ 2 mm ($n = 4$). Particle size distribution was determined by sieve and pipette technique according to DIN ISO 11277 (2002). For the determination of the pH value, composite samples were taken systematically from the plot and mixed into one sample. The pH values were measured in 0.01 M CaCl₂ solution (1:2.5 soil:CaCl₂) according to DIN ISO 11260 (2018). The contents of total carbon and nitrogen were determined with a CNS Element-Analyzer (Vario EL III, Elementar, Langenselbold/Germany) according to DIN ISO 10694 (1995) and DIN ISO 13878 (1998). Inorganic carbon contents were determined from the CO₂ release during treatment with 10 % HCl (Scheibler method, (DIN ISO 10693, 1997)). The organic carbon content was determined from the difference between the results of the total carbon content and carbonate carbon content. Organic carbon content was converted to soil organic matter content using the factor 1.72.

Clay mineralogy was analyzed using X-ray diffraction (XRD) (D2 Phaser Bruker AXS GmbH, Karlsruhe/Germany). The analysis required a separation of clay from the remaining soil by disaggregation and dispersion. Then, the homogenized clay paste was prepared on flat glass discs and measured in three steps. First, the air-dried sample was analyzed in XRD, second, the sample was vaporized with ethylene glycol and analyzed in XRD and third, the sample was dried at 550°C and analyzed again (Moore and Reynolds, 1997). The steps were followed by a graphical analysis of the clay minerals with the software DIFFRAC.EVA version V4 (Bruker AXS GmbH, Karlsruhe/Germany).

6.3.3 Measurements on undisturbed soil cores

Saturated hydraulic conductivity (k_s) was measured on undisturbed soil cores (100 cm³) based on the falling-head method as described in Hartge and Horn (2009). The pore size distribution was obtained from undisturbed soil cores (100 cm³) by successively draining initially water-saturated samples to matric potentials of $\psi_m = -6$ and -30 kPa using ceramic plates under vacuum. To derive the fine pore percentage ($\emptyset < 0.2$ μm), small soil cores (2 cm³) were drained to $\psi_m = -1500$ kPa in an air

pressure device (Hartge and Horn, 2009). Bulk density (ρ_B) was determined from the weight to volume ratio after oven-drying at 105°C for 24 h. Total porosity (TP) was determined from ρ_B and solid phase density of quartz (ρ_s) (eq. 6-2).

$$TP = \left(1 - \frac{\rho_B}{\rho_s}\right) \cdot 100 \quad \text{eq. 6-2}$$

with

TP = total porosity (vol.-%)

ρ_B = bulk density ($g\ cm^{-3}$)

ρ_s = solid phase density of quartz ($2.65\ g\ cm^{-3}$)

The percentage of wide coarse pores $\varnothing > 50\ \mu m$ was determined from the difference between total porosity and the volumetric water content at -6 kPa. The percentage of narrow coarse pores ($\varnothing 50-10\ \mu m$) was determined from the difference in volumetric water content at -6 kPa and -30 kPa. The percentage of medium pores ($\varnothing 10-0.2\ \mu m$) was determined from the difference in volumetric water contents at -30 kPa and -1500 kPa.

Air conductivity (k_a) was measured on pre-drained soil samples at $\psi_m = -6$ and -30 kPa with an air permeameter under steady-state flow conditions (eq. 6-3) (Peth, 2004).

$$k_a = \frac{\rho \cdot g \cdot q \cdot L_s}{A_s \cdot p} \quad \text{eq. 6-3}$$

with

k_a = air conductivity ($cm\ s^{-1}$)

ρ = air density ($kg\ m^{-3}$)

g = gravitational acceleration ($m\ s^{-1}$)

q = volumetric flow rate ($m^3\ s^{-1}$)

L_s = length of the sample (m)

A_s = surface of the sample (m^2)

p = pneumatic pressure (Pa)

Shear strength was measured on undisturbed soil cores ($236\ cm^3$) at $\psi_m = -6$ kPa in a drained uniaxial confined compression test (direct shear test) at defined loads: 20, 50, 100, 200 and 300 kPa. The shear strength (τ) parameters cohesion (c) and angle

of internal friction (φ) are defined by the Mohr Coulomb failure line (Kézdi and Rétháti, 1988) (eq. 6-4):

$$\tau = \sigma_n \cdot \tan(\varphi) + c \quad \text{eq. 6-4}$$

with

τ = shear strength (kPa)

φ = angle of internal friction (rad)

c = cohesion (kPa)

σ_n = normal stress (kPa)

6.3.4 Measurements on individual aggregates

The tensile strength (Y) of soil aggregates ($\varnothing \sim 16$ mm) was derived from crushing tests. For this purpose, undisturbed soil blocks (1 dm³) were extracted in the field and the aggregates were collected from the field blocks in the laboratory by breaking them along their weakest rupture planes. The collected aggregates were subsequently air-dried in preparation for the crushing test. A second set of aggregates was collected from undisturbed soil cores (236 cm³), which had been pre-drained to $\psi_m = -30$ kPa using a vacuum plate. The aggregates were collected from the pre-drained soil cores by breaking them along their weakest rupture planes and exposed to the crushing test immediately afterwards. In the crushing test, every aggregate ($n = 20$) was crushed separately by applying a vertical compressive force using an electromechanical test system (Instron 5569, Darmstadt/Germany, software Merlin 5.31.00) at a constant rate of displacement of 5 mm min⁻¹. The tensile strength (Y) was calculated according to Dexter and Kroesbergen (1985) (eq. 6-5):

$$Y = 0.576 \cdot F / d^2 \quad \text{eq. 6-5}$$

with

Y = tensile strength of aggregate (kPa)

F = polar force at failure (N)

d = aggregate diameter (mm)

6.3.5 Statistical analysis

The statistical analysis was carried out using the statistical software R version 3.5.1 (R Core Team, 2017). Based on graphical residual analysis, the values of pH and the water retention parameters were assumed to be normally distributed and heteroscedastic due to different lime application rates at the site. The values of k_s and k_a were not normally distributed. Based on these assumptions, an analysis of variances (One-Factor ANOVA) was performed (Snedecor and Cochran, 1996). All values were evaluated comparing the means of the factor levels treatment and plot for each site separately (Tukey-Test). The significance of the different tests was set at an α -Level of 5 %. Linear relationships between selected parameters of interest were expressed by Pearson's correlation coefficient (r). Box-Whisker plots were chosen for the graphical presentation, showing median (mid-line), 25th/75th percentile (lower/upper border of the box), 95th and 5th percentile (Whiskers), 1st and 99th percentile (lower/upper line), outliers (dots) and mean (squares) (McGill et al., 1978).

6.4 Results

6.4.1 Soil characteristics

Soil texture, organic matter content and information on the clay mineralogy of the three sites are summed up in Table 6-2. In Neubrandenburg, the topsoil (0–30 cm depth) is a loamy sand with a clay content of 11 % and a soil organic matter content of 13 g kg⁻¹ (Frank et al., 2019). The dominant clay mineral is the 1:1 mineral nacrite (polytypic with kaolinite). In Barlt, the topsoil (0–30 cm depth) is a silty clay with a clay content of 45 % and a soil organic matter content of 56 g kg⁻¹ (Frank et al., 2020). The dominant clay minerals are the 2:1 minerals montmorillonite and illite. In Schrecksbach, the topsoil (0–30 cm depth) is a clayey silt with a clay content of 24 % and a soil organic matter content of 26 g kg⁻¹. The dominant clay mineral is the 1:1 mineral kaolinite.

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Table 6-2: Soil organic matter content [g kg⁻¹], particle size distribution [%] and dominant clay minerals in the topsoil (0–30 cm depth) from the control plots (Lime0) at the three study sites Neubrandenburg, Barlt and Schrecksbach.

		Neubrandenburg	Barlt	Schrecksbach
Soil organic matter	[g kg ⁻¹]	13	56	26
Particle size distribution	[%]			
	Sand	64	5	7
	Silt	25	50	69
	Clay	11	45	24
Dominant clay minerals		nacrite	montmorillonite & illite	kaolinite

6.4.2 pH values

At all three sites, the pH in the Lime1 and Lime1.5 plots was at the recommended pH-level 12 and 24 months after lime application (Frank et al., 2019, 2020) (Table 6-3).

Table 6-3: pH (CaCl₂ (0.01 M)) for the lime (CaCO₃) treatments (Lime0 = control, Lime1 = lime recommendation according to VDLUFA (2000a) Lime1.5 = lime recommendation according to VDLUFA x 1.5) at the study sites Neubrandenburg, Barlt and Schrecksbach (12 and 24 months after lime application). Values are presented as arithmetic means. n = 2

Site	Treatment	12 months after lime application pH [-]	24 months after lime application pH [-]
Neubrandenburg	Lime0	6.19	6.15
	Lime1	6.84	6.63
	Lime1.5	6.93	6.74
Barlt	Lime0	5.55	5.91
	Lime1	6.22	6.23
	Lime1.5	6.28	6.46
Schrecksbach	Lime0	5.45	5.76
	Lime1	6.47	6.92
	Lime1.5	6.86	6.96

6.4.3 Pore size distribution and conductivities

At the study site in Neubrandenburg, no significant differences were found for wide coarse pores on limed plots compared to the control plots 12 and 24 months after lime application (Fig. 6-1). The percentage of narrow coarse pores was significantly lower on limed plots (Lime1) compared to control plots (Lime0) ($p < 0.05$) 12 months after lime application, while no changes were obtained 24 months after lime application and with tillage between the two sampling times. The percentage of medium pores was significantly lower on limed plots (Lime1.5) compared to control plots (Lime0) ($p < 0.05$) 12 months after lime application, while no changes were obtained 24 months after lime application.

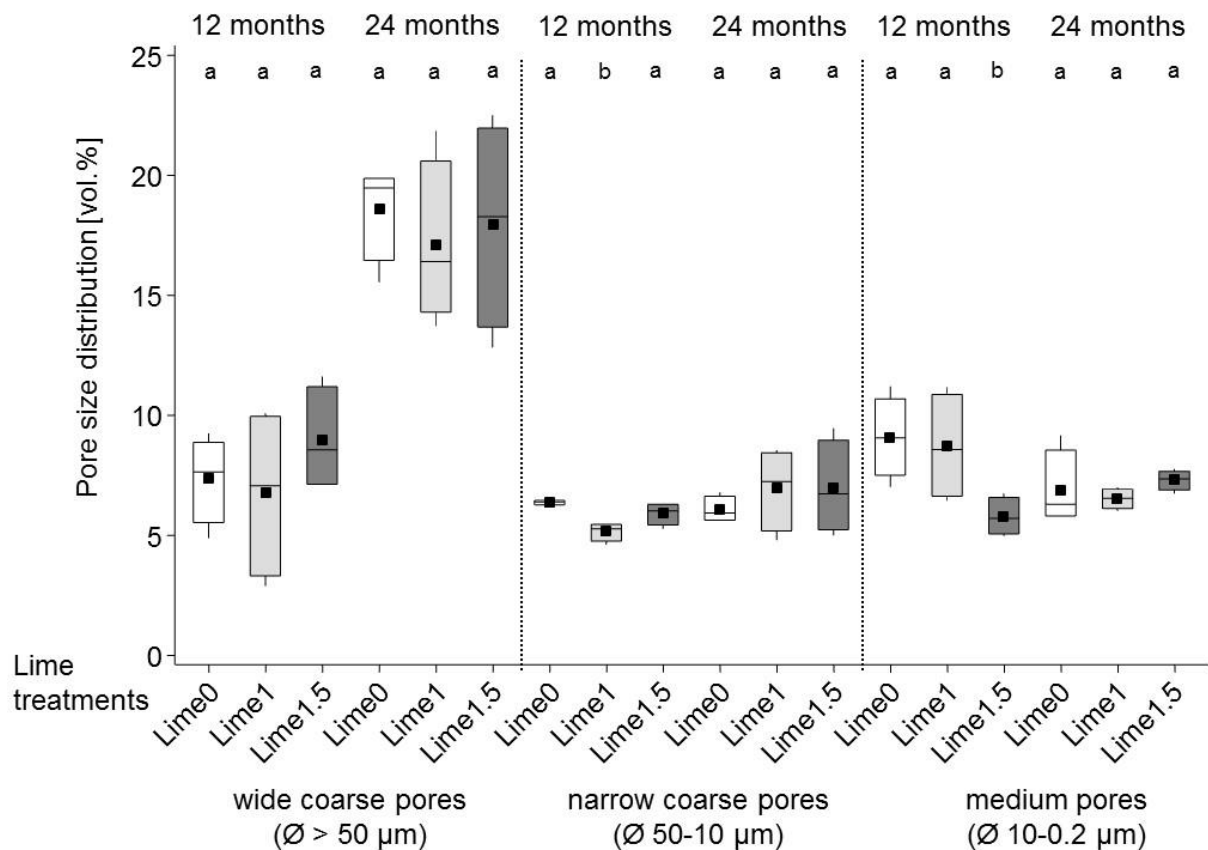


Fig. 6-1: Proportion of pore size class [vol.-%] of wide coarse pores, narrow coarse pores and medium pores at the study site Neubrandenburg obtained from the topsoil of the lime treatments (Lime0: control, Lime1: lime recommendation according to VDLUFA (2000a), Lime1.5: lime recommendation according to VDLUFA (2000a) x 1.5) 12 and 24 months after lime application ($n = 12$). Squares denote arithmetic means. b (a) indicates treatment (not) significantly different from the control (Lime0) at $p < 0.05$.

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At the study site in Barlt, no significant changes were found for the wide coarse pores and narrow coarse pores on limed plots compared to the control (Fig. 6-2). The percentage of medium pores was significantly lower on Lime1.5 plots compared to the control (Lime0) ($p < 0.05$) 12 months after lime application, while, no significant changes were obtained 24 months after lime application.

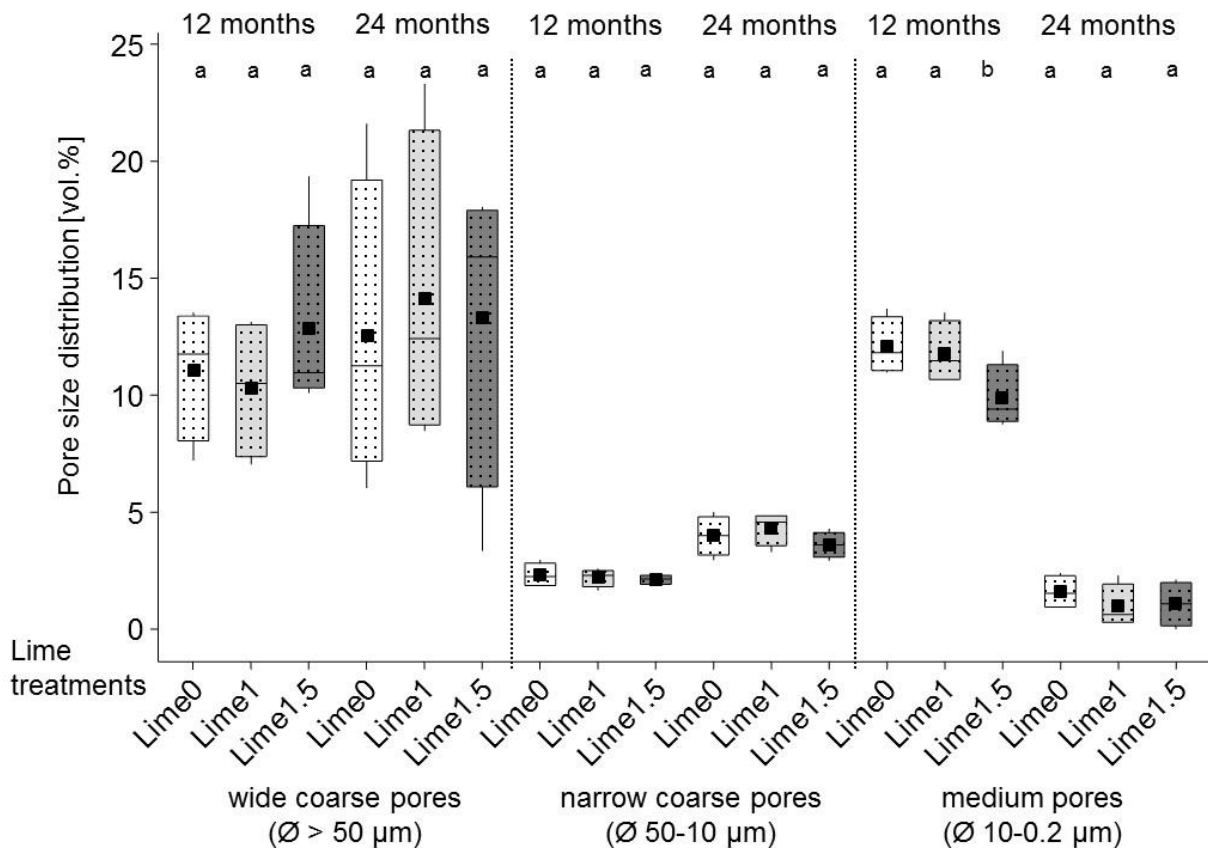


Fig. 6-2: Proportion of pore size class [vol.-%] of wide coarse pores, narrow coarse pores and medium pores at the study site Barlt obtained from the topsoil of the lime treatments (Lime0: control, Lime1: lime recommendation according to VDLUFA (2000a), Lime1.5: lime recommendation according to VDLUFA (2000a) x 1.5) 12 and 24 months after lime application ($n = 12$). Squares denote arithmetic means. b (a) indicates treatment (not) significantly different from the control (Lime0) at $p < 0.05$.

At the study site in Schrecksbach, no significant changes were found for the wide coarse pores and narrow coarse pores on limed plots compared to the control, with tillage between the two sampling times (Fig. 6-3). The percentage of medium pores was significantly lower on limed plots (Lime1 and Lime1.5) compared to the control (Lime0) ($p < 0.05$) 12 months after lime application, while, the percentage of medium pores was significantly lower only on Lime1.5 plots compared to the control (Lime0) ($p < 0.05$) 24 months after lime application.

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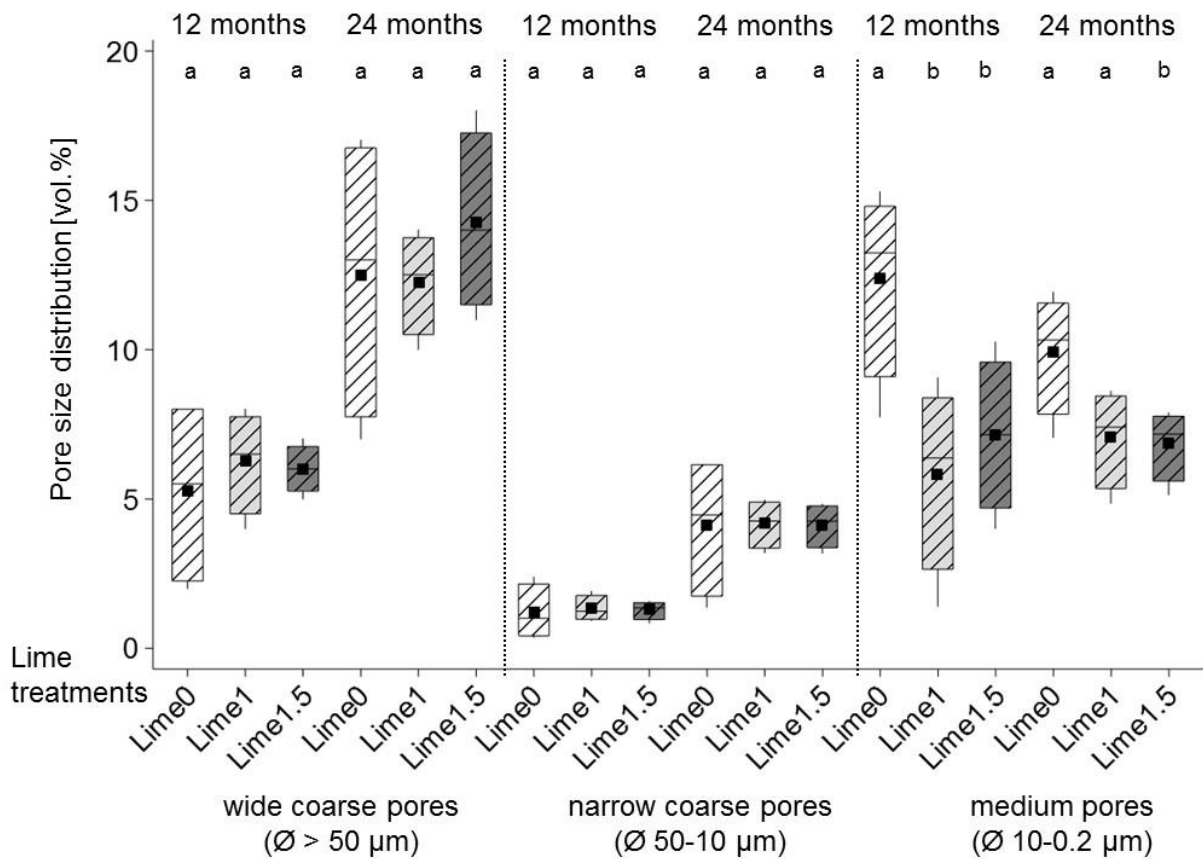


Fig. 6-3: Proportion of pore size class [vol.-%] of wide coarse pores, narrow coarse pores and medium pores at the study site Schrecksbach obtained from the topsoil of the lime treatments (Lime0: control, Lime1: lime recommendation according to VDLUFA (2000a), Lime1.5: lime recommendation according to VDLUFA (2000a) x 1.5) 12 and 24 months after lime application (n = 12). Squares denote arithmetic means. b (a) indicates treatment (not) significantly different from the control (Lime0) at p < 0.05.

The saturated hydraulic conductivity (k_s) was significantly higher in the limed plots compared to the control plots ($p < 0.05$) 12 months after lime application at the study site in Neubrandenburg, as already shown in Frank et al. (2019). Air conductivity (k_a) at $\psi_m = -30$ kPa correlated significantly with k_s for the Lime0 treatment ($r = 0.99$, $p = 0.04$) and for the Lime1.5 treatment ($r = 0.95$, $p = 0.05$) at the study site in Neubrandenburg 12 months after the lime application (Fig. 6-4). However, there were no significant correlations between k_a and k_s at Barlt and Schrecksbach 12 months after lime application.

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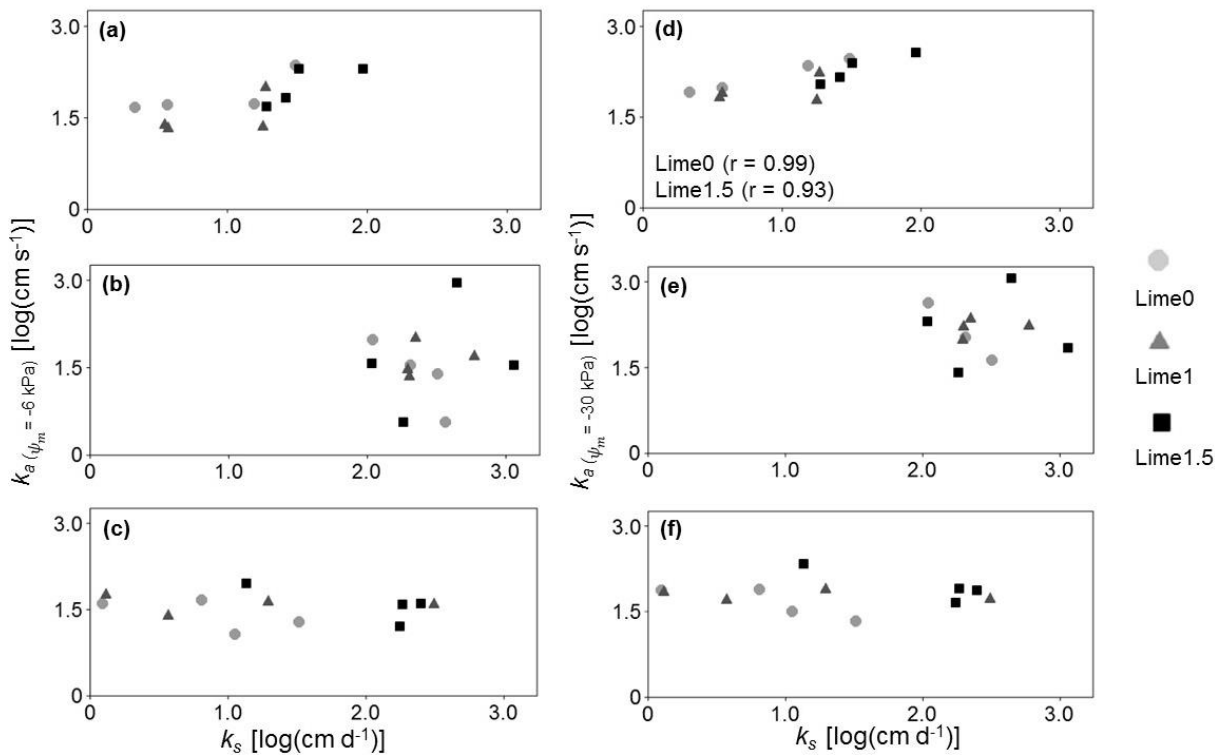


Fig. 6-4: Air conductivity k_a [cm s⁻¹] at $\psi_m = -6$ kPa (logarithmic displays) as a function of saturated hydraulic conductivity k_s [cm d⁻¹] (logarithmic displays) (left) at: Neubrandenburg (a), Barlt (b) and Schrecksbach (c) and at $\psi_m = -30$ kPa (logarithmic displays) (right) as a function of saturated hydraulic conductivity k_s [cm d⁻¹] (logarithmic displays) at: Neubrandenburg (d), Barlt (e) and Schrecksbach (f) with the lime treatments (Lime0: control, Lime1: lime recommendation according to VDLUFA (2000a), Lime1.5: lime recommendation according to VDLUFA (2000a) x 1.5) 12 months after lime application. Values are presented as geometric means ($n = 12$). Pearson's correlation coefficient (r) is given for the lime treatments with significant correlations ($p < 0.05$).

The saturated hydraulic conductivity (k_s) was significantly higher in the limed plots compared to the control plots ($p < 0.05$) 24 months after lime application at the study site in Barlt, as already shown in Frank et al. (2020). None of the correlations between air conductivity (k_a at $\psi_m = -6$ kPa and at $\psi_m = -30$ kPa) and k_s were significant ($p < 0.05$) 24 months after lime application (Fig. 6-5).

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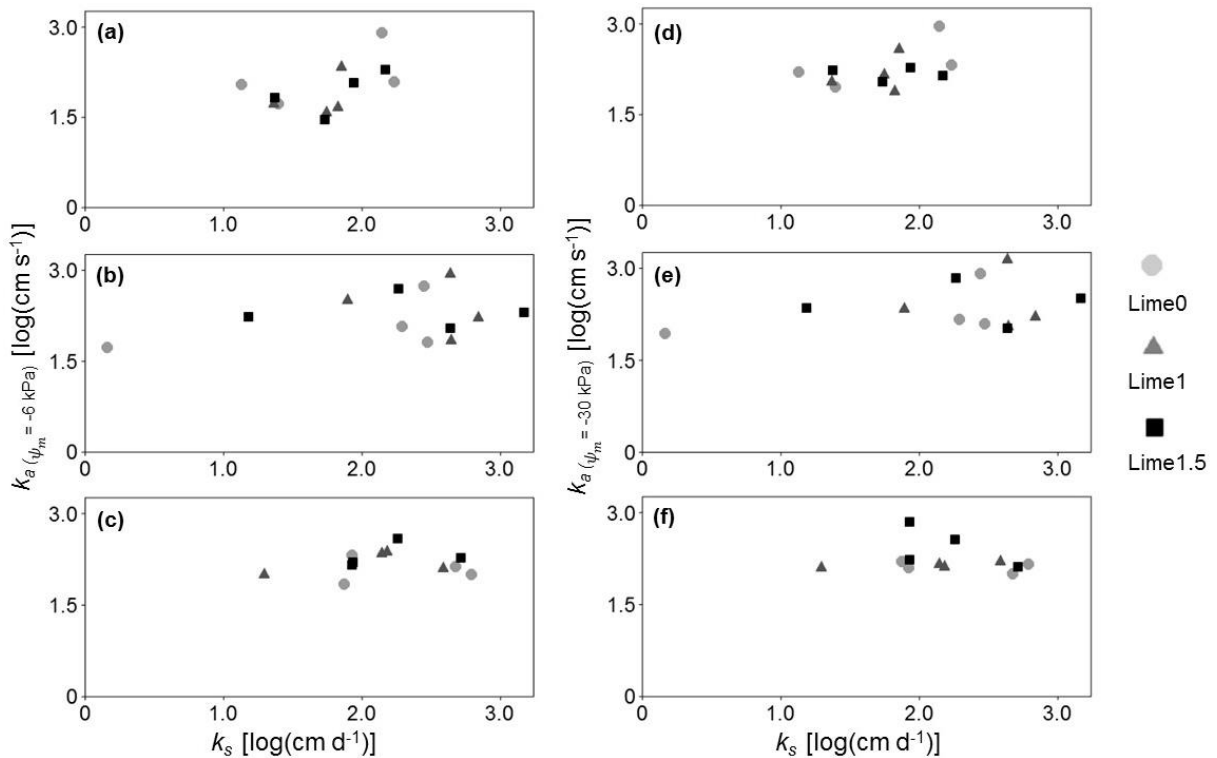


Fig. 6-5: Air conductivity k_a [cm s⁻¹] at $\psi_m = -6$ kPa (logarithmic displays) as a function of saturated hydraulic conductivity k_s [cm d⁻¹] (logarithmic displays) (left) at: Neubrandenburg (a), Barlt (b) and Schrecksbach (c) and at $\psi_m = -30$ kPa (logarithmic displays) (right) as a function of saturated hydraulic conductivity k_s [cm d⁻¹] (logarithmic displays) at: Neubrandenburg (d), Barlt (e) and Schrecksbach (f) with the lime treatments (Lime0: control, Lime1: lime recommendation according to VDLUFA (2000a), Lime1.5: lime recommendation according to VDLUFA (2000a) x 1.5) 24 months after lime application. Values are presented as geometric means ($n = 12$). Pearson's correlation coefficient (r) is given for the lime treatments with significant correlations ($p < 0.05$).

Air conductivity (k_a) at $\psi_m = -6$ kPa significantly correlated with the total porosity ($r = 0.99$, $p = 0.006$) and with the wide coarse pores ($r = 0.94$, $p = 0.04$) at the lime 1.5 treatment for the study site in Neubrandenburg 12 months after lime application (Fig. 6-6). The correlation between k_a and the total porosity ($r = 0.93$, $p = 0.007$) significantly correlated 24 months after the application of lime at Lime1 treatments, as well as the correlation between k_a and the percentage of the wide coarse pores significantly correlated ($r = 0.96$, $p = 0.04$) at the Lime1 treatments 24 months after lime application.

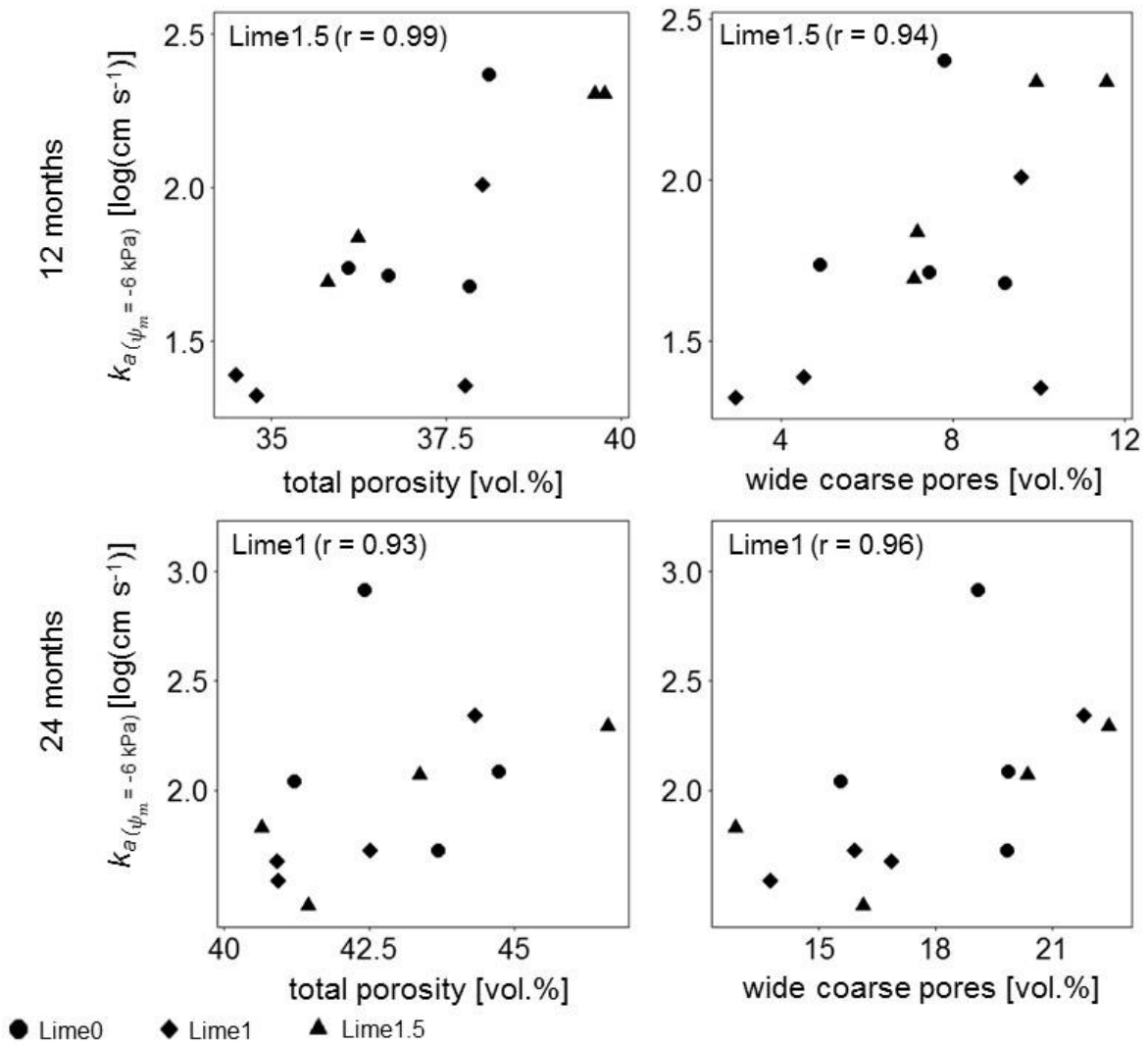


Fig. 6-6: Air conductivity k_a [cm s⁻¹] at $\psi_m = -6$ kPa (logarithmic display) as a function of the total porosity [vol.%] and the wide coarse pores [vol.%] at the study site Neubrandenburg with the lime treatments (Lime0: control, Lime1: lime recommendation according to VDLUFA (2000a) Lime1.5: lime recommendation according to VDLUFA (2000a) x 1.5) 12 and 24 months after lime application. Values are presented as geometric means ($n = 12$). Pearson's correlation coefficient (r) is given for the lime treatments with significant correlations ($p < 0.05$).

Air conductivity (k_a) at $\psi_m = -6$ kPa significantly correlated with total porosity ($r = 0.99$, $p = 0.01$) at the Lime1.5 treatments for the study site in Barlt 12 months after lime application (Fig. 6-7). While the correlation between k_a and the total porosity ($r = 0.95$,

$p = 0.05$) as well as k_a with the wide coarse pores ($r = 0.98$, $p = 0.02$) significantly correlated on Lime0 treatments 24 months after the application of lime.

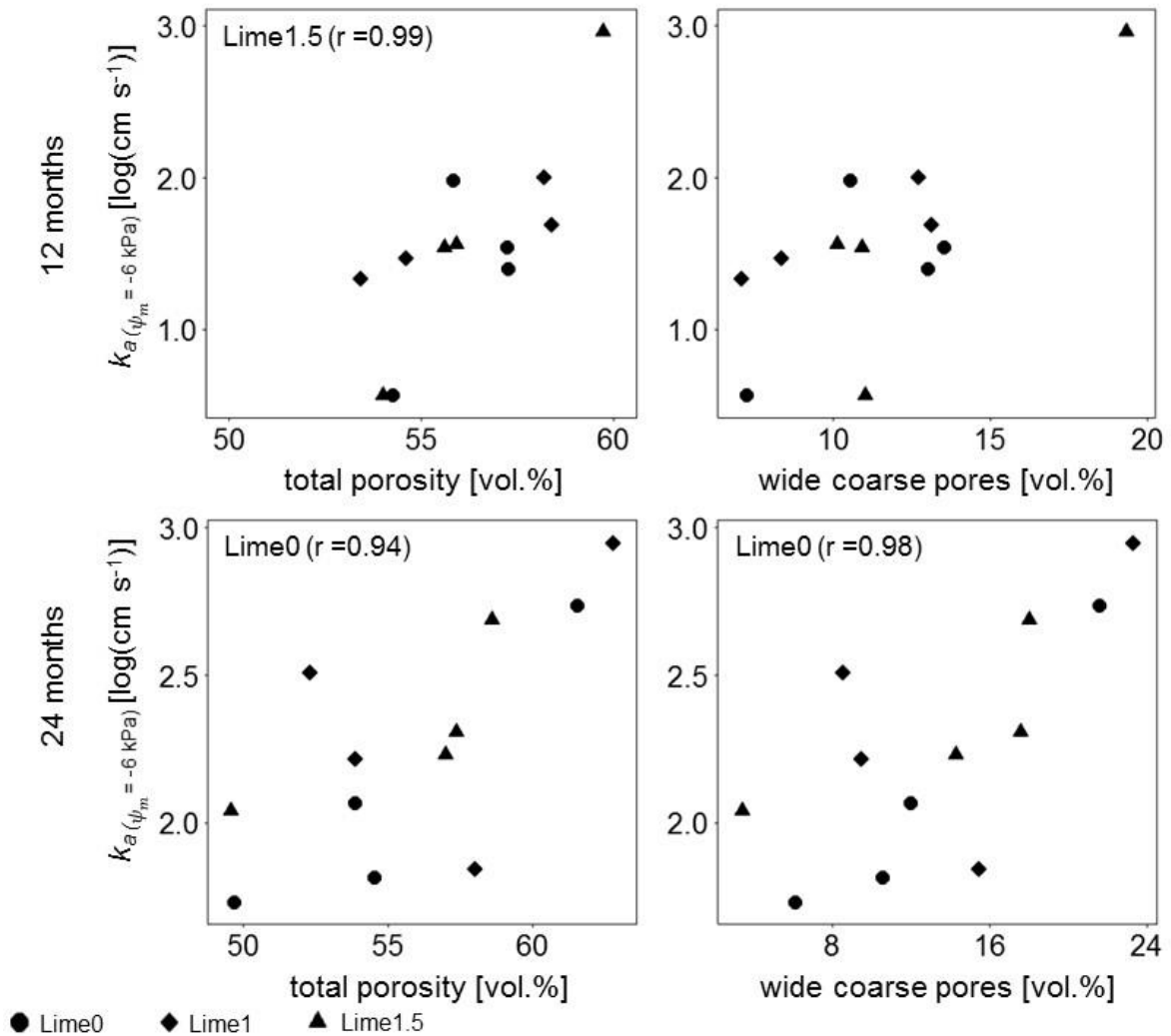


Fig. 6-7: Air conductivity k_a [cm s^{-1}] at $\psi_m = -6$ kPa (logarithmic display) as a function of the total porosity [vol.%] and the wide coarse pores [vol.%] at the study site Barlt with the lime treatments (Lime0: control, Lime1: lime recommendation according to VDLUFA (2000a) Lime1.5: lime recommendation according to VDLUFA (2000a) $\times 1.5$) 12 and 24 months after lime application. Values are presented as geometric means ($n = 12$). Pearson's correlation coefficient (r) is given for the lime treatments with significant correlations ($p < 0.05$).

At the study site in Schrecksbach, air conductivity (k_a) at $\psi_m = -6$ kPa significantly correlated with the percentage of the total porosity ($r = 0.99$, $p = 0.01$) at the Lime1 treatments 12 both after lime application. No further correlations were determined for the different lime treatments both 12 and 24 months after lime application (Fig. 6-8).

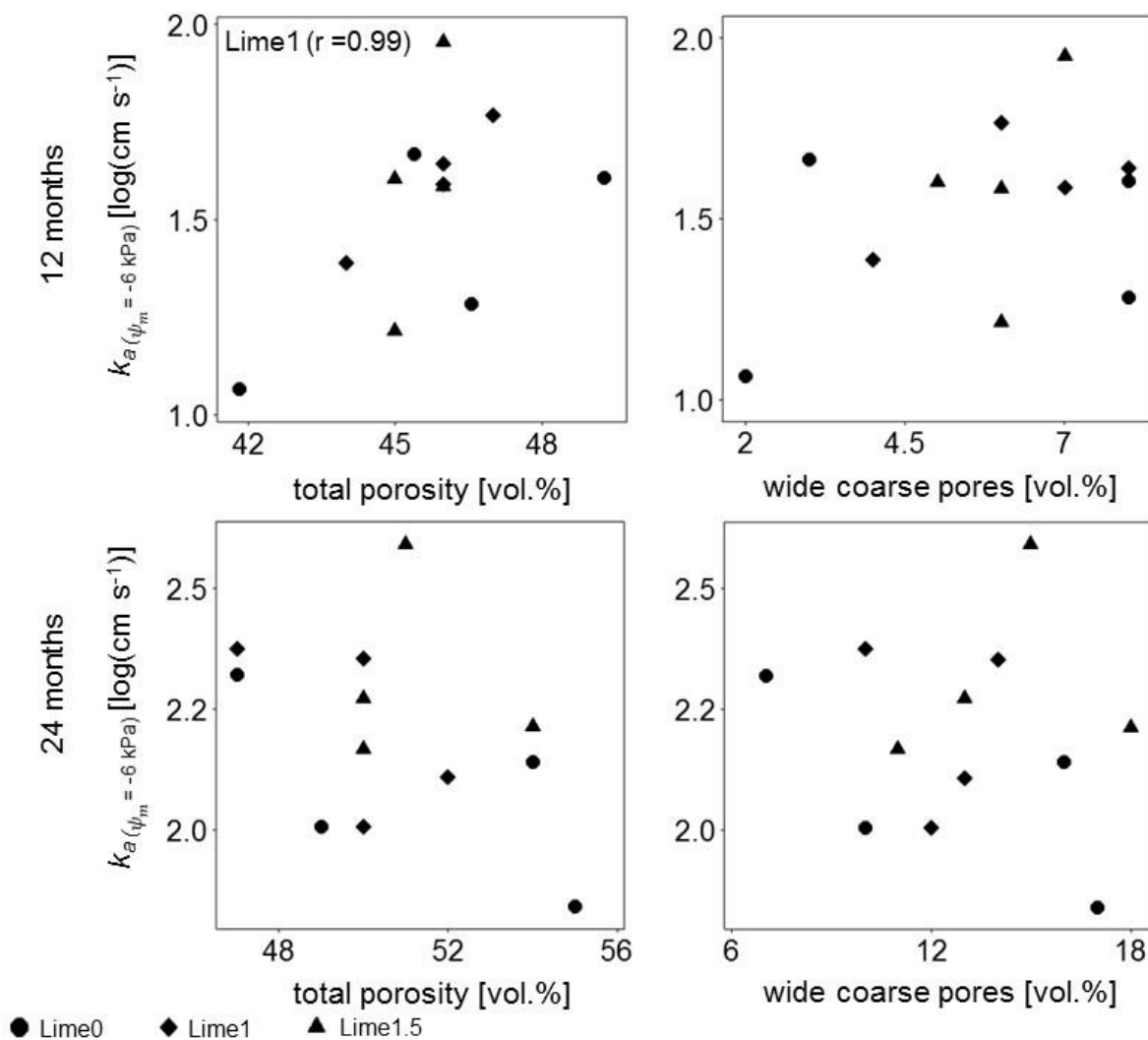


Fig. 6-8: Air conductivity k_a [cm s^{-1}] at $\psi_m = -6$ kPa (logarithmic display) as a function of the total porosity [vol.%] and the wide coarse pores [vol.%] at the study site Schrecksbach with the lime treatments (Lime0: control, Lime1: lime recommendation according to VDLUFA (2000a), Lime1.5: lime recommendation according to VDLUFA x 1.5) 12 and 24 months after lime application. Values are presented as geometric ($n = 12$). Pearson's correlation coefficient (r) is given for the lime treatments with significant correlations ($p < 0.05$).

6.4.4 Shear parameters and shear strength

In Neubrandenburg, a higher cohesion (c) in limed treatments compared to the control was determined 12 months after lime application. However, the differences were not significant (Table 6-4). 24 months after lime application, with tillage between, c was lower and ϕ was higher in the limed plots compared to the control.

In Barlt, c is significantly higher and ϕ is significantly lower on the limed plots (Lime1 and Lime1.5) compared to the control 12 months after the lime application (Table 6-4). A (not significantly) higher c with an unchanged ϕ in limed plots compared to the control

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plots occurred 24 months after lime application with tillage between the two sampling times.

In contrast to the former sites, no significant changes were determined for c in the limed plots (Lime1 and Lime1.5) compared to the control (Lime0) at the study site in Schrecksbach throughout the whole period (Table 6-4). The ϕ was higher in limed plots compared to the control plots 12 months after lime application.

Table 6-4: Cohesion (c) in kPa and angle of internal friction (ϕ) in ° at $\psi_m = -6$ kPa in the topsoil of the lime treatments (Lime0: control, Lime1: lime recommendation according to VDLUFA (2000a), Lime1.5: lime recommendation according to VDLUFA x 1.5) at the three study sites Neubrandenburg, Barlt and Schrecksbach 12 and 24 months after lime application and incorporation. Values are presented as arithmetic means. Asterisks indicate treatments significantly different from control (Lime0) at $p < 0.05$, ($n = 2$).

Treatment	Neubrandenburg		Barlt		Schrecksbach	
	c kPa	ϕ (°)	c kPa	ϕ (°)	c kPa	ϕ (°)
	12 months after lime application					
Lime0	19	37	9	37	27	33
Lime1	22	34	25*	29*	31	34
Lime1.5	23	35	33*	27*	22	36
	24 months after lime application					
Lime0	17	34	26	32	27	32
Lime1	13	38	30	32	23	33
Lime1.5	15	37	32	32	23	33

Maximum shear strength (τ) measured as a function of normal stress (σ_n) was higher on the limed plots (Lime1 and Lime1.5) compared to the control (Lime0) 12 months after lime application for the study site in Schrecksbach (Fig. 6-9b), while on the study site in Barlt, τ was highest on the Lime0 plots (Fig. 6-9c). 24 months after lime application, higher τ occurred on the limed plots (Lime1 and Lime1.5) compared to the control (Lime0) at the study sites Neubrandenburg (Fig. 6-9d) and Schrecksbach (Fig. 6-9e), while no changes were obtained for the study site in Barlt (Fig. 6-9f).

Effect of CaCO_3 application on mechanical strength and hydraulic properties
in arable soils

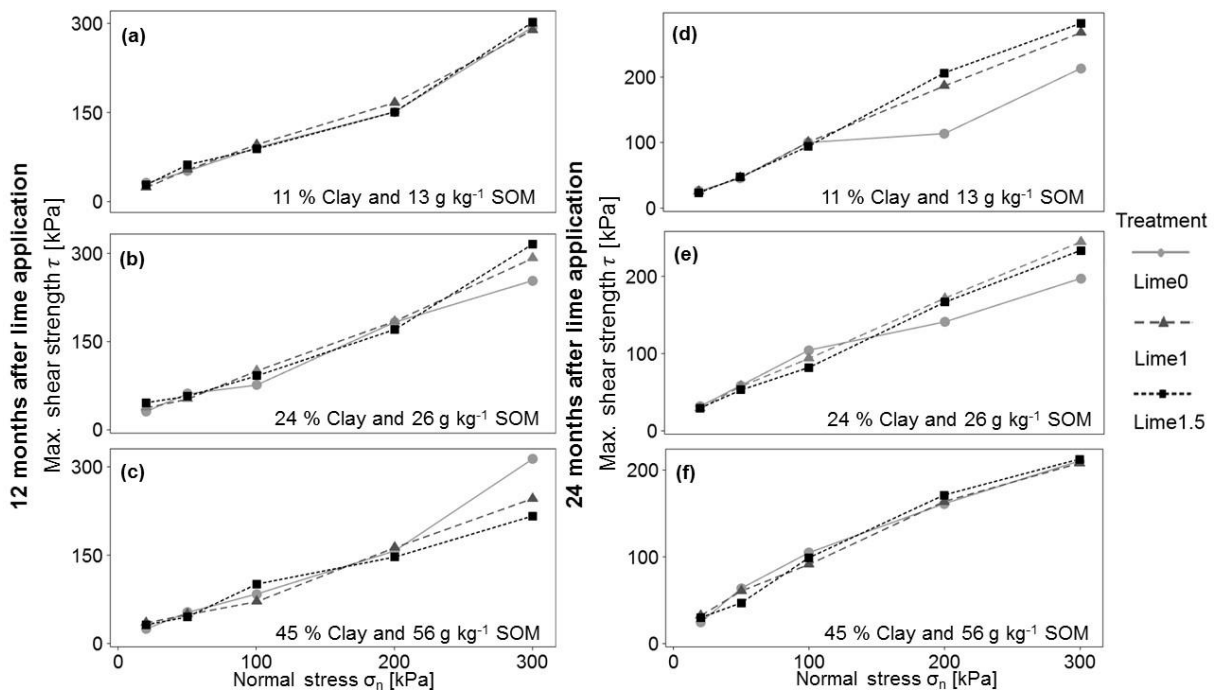


Fig. 6-9: Maximum shear strength (τ) [kPa] as a function of normal stress (σ_n) (20, 50, 100, 200 and 300 kPa) from the topsoil of the lime treatments (Lime0: control, Lime1: lime recommendation according to VDLUFA (2000a), Lime1.5: lime recommendation according to VDLUFA (2000a) x 1.5) 12 months after lime application at: Neubrandenburg (a), Schrecksbach (b) and Barlt (c) and 24 months after lime application at: Neubrandenburg (d), Schrecksbach (e) and Barlt (f) (n = 4).

6.4.5 Aggregate tensile strength

At all three sites, no significant differences were determined for the tensile strength (Y) of air-dried aggregates 12 months after lime application (Fig. 6-10), although often Y increased with increasing lime treatment (Lime1 and Lime1.5) compared to the control (Lime0).

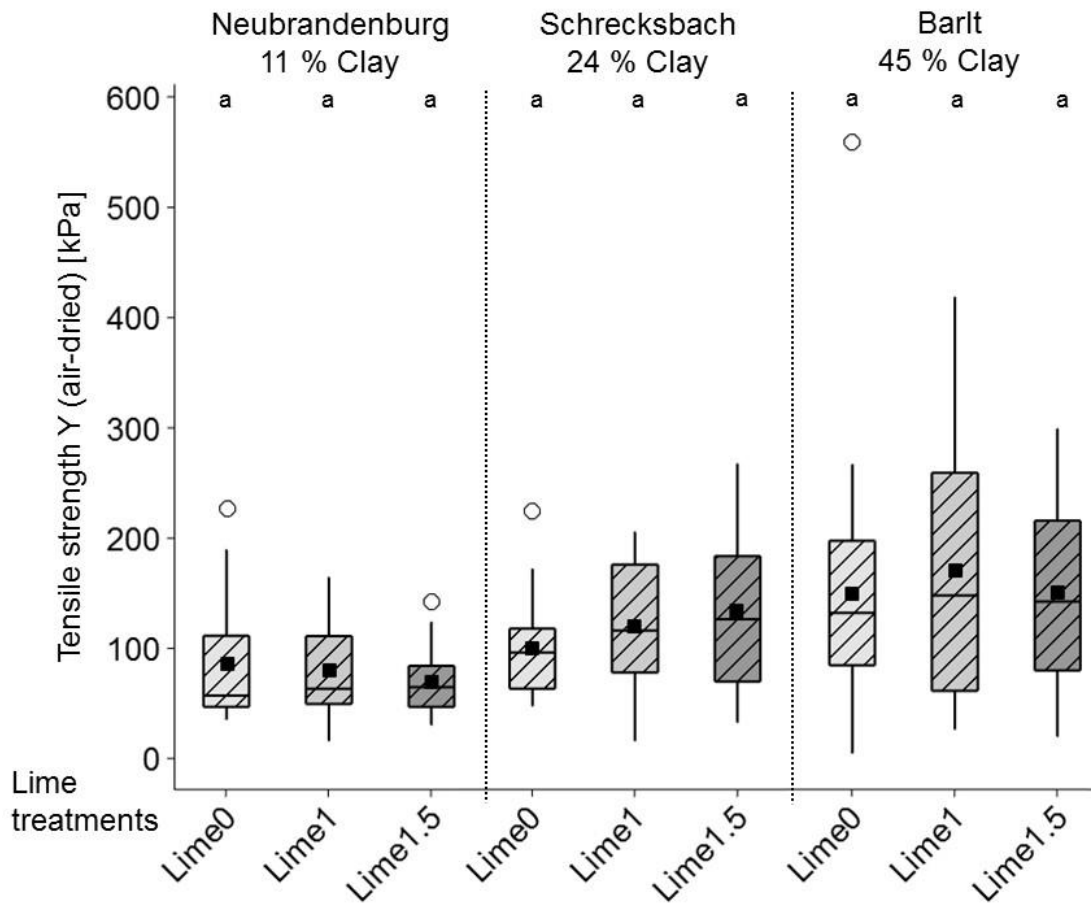


Fig. 6-10: Tensile strength (Y, in kPa) of air-dried aggregates (diameter 16 mm) from the topsoil of the lime treatments (Lime0: control, Lime1: lime recommendation according to VDLUFA (2000a), Lime1.5: lime recommendation according to VDLUFA x 1.5) 12 months after lime application at the three survey sites Neubrandenburg, Schrecksbach and Barlt (n = 20). Squares denote arithmetic means. b (a) indicates treatment (not) significantly different from the control (Lime0) at p < 0.05.

Only the pre-drained soil aggregates ($\psi_m = -30$ kPa) from the study site in Barlt showed significant increases of Y in the Lime1.5 treatment compared to the control (Lime0) 24 months after lime application and incorporation (Fig. 6-11).

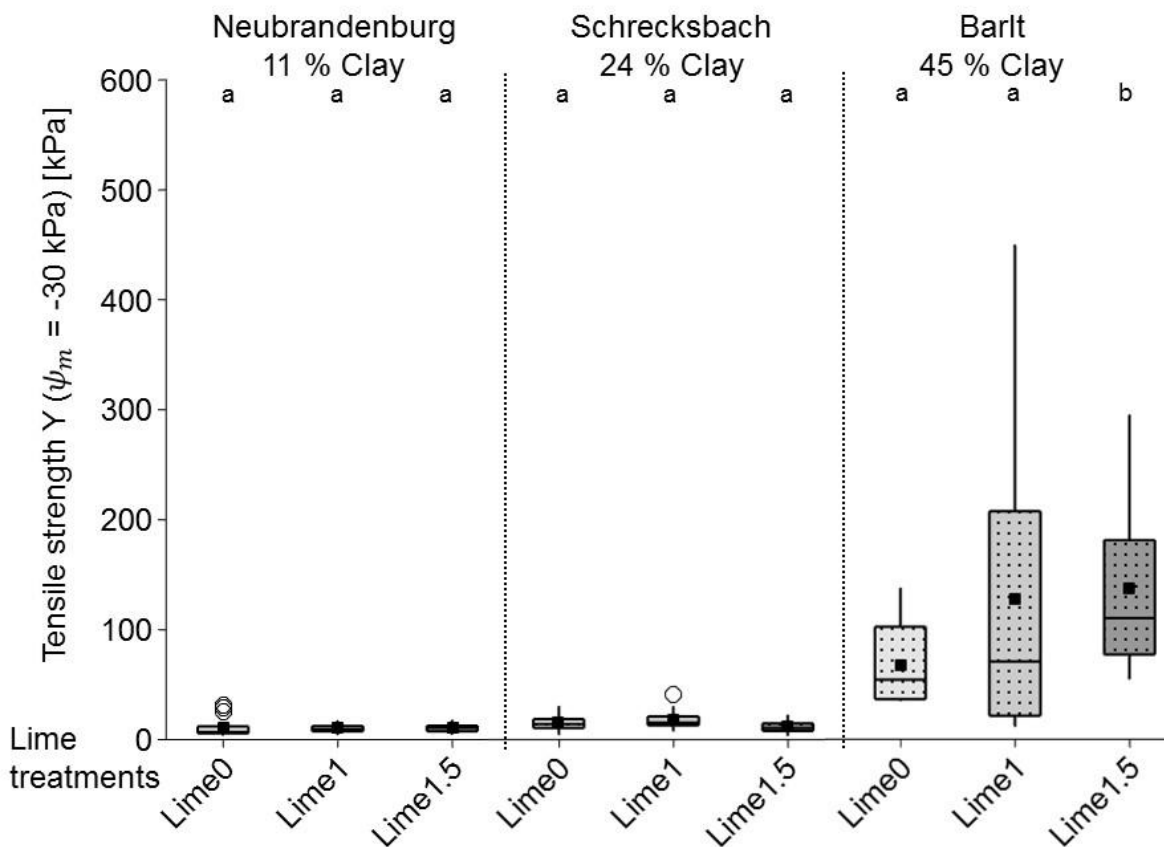


Fig. 6-11: Tensile strength (Y , in kPa) of pre-drained ($\psi_m = -30$ kPa) aggregates (diameter 16 mm) from the topsoil of the lime treatments (Lime0: control, Lime1: lime recommendation according to VDLUFA (2000a), Lime1.5: lime recommendation according to VDLUFA x 1.5) 24 months after lime application at the three survey sites Neubrandenburg, Schrecksbach and Barlt ($n = 20$). Squares denote arithmetic means. b (a) indicates treatment (not) significantly different from the control (Lime0) at $p < 0.05$.

6.5 Discussion

Arable soils have to withstand mechanical stresses induced by agricultural machineries in order to maintain a high physical but also chemical and biological soil quality, as defined by a structural state that balances transport and storage of water and nutrients and guarantees optimal living conditions for flora and fauna. In our research, the soils at the three field trials have different soil organic matter (SOM) and clay content (Table 6-2), and are therefore very suitable for determining the influence of lime in different soils. The air conductivity, air capacity (pores $\varnothing > 50 \mu\text{m}$), total porosity as well as the saturated hydraulic conductivity are useful indicators of physical soil quality, because they respond to changes in land management (Reynolds et al., 2007). In addition, the shear resistance, determined by the cohesion and angle of internal friction, as well as aggregate tensile strength allow predictions regarding the

quality of the soil structure and individual aggregates (Santos et al., 1997; Urbanek et al., 2014), and how these parameters change under the influence of management measures, e.g. a single addition of lime (Bohne et al., 1985; Hartge and Ellies, 1977; Zhang and Hartge, 1990).

6.5.1 Changes of pH values due to lime application

It is well known that acidic soils can be ameliorated by the application of lime in order to improve crop production (Goulding, 2016). The pH value of the soil is subjected to fluctuations and changes over time, which in turn can influence the effect of a single lime application. Under humid climatic conditions, soil acidification occurs gradually due to continuous input or formation of acids. Due to the high specific surface area of clay, a large proportion of reactive binding sites are available for the uptake of cations and organic components. Thus, when lime in the form of CaCO₃ is applied, the dispersion of clay particles is minimized by the increase in negative charges as a result of the increase in soil pH over time after application. Here it is crucial that especially the polyvalent cations Ca²⁺ and Mg²⁺ adsorb at the exchange sites (Frank et al., 2020). These chemical changes in the soil solution also show effects on the physical soil parameters already 12 months after liming, as discussed in the next section.

6.5.2 Alterations of the pore systems by liming

Lime application, in form of CaCO₃, and incorporation into the soil induces a release of Ca²⁺ ions. The Ca²⁺ in the soil solution compresses the diffuse double layer and promotes flocculation of the (clay) mineral particles (Haynes and Naidu, 1998). Furthermore, Ca²⁺ acts as a bridge between organic substances and (clay) mineral particles, and the formation of secondary carbonates creates bridges between mineral particles and causes cementation over time. A shift in the pore ratios, including secondary, large, interaggregate pores (wide coarse pores, $\varnothing > 50 \mu\text{m}$), is the result. Similar to the findings of Kirkham et al. (2007), who explained the increase in k_s after liming by a shift in pore size distribution to more medium and coarse pores, we observed a rearrangement in the pore size distribution with a shift from the wide coarse pores to a higher percentage of narrow coarse pores 24 months after liming for the sandy soil (Fig. 6-1). The plant-available water stored in these pore sizes may improve

the plant growth at our study site, especially in dry periods. Gas and water transport will occur preferentially through the inter-aggregate pore system formed by macropores (Horn and Smucker, 2005). Thus, a higher percentage of coarse pores, combined with a lower proportion of medium pores (Fig. 6-2), as observed in the clay soil (study site Barlt), can lead to an increase in gas and water flow, as long as no tillage disrupts the soil structure. The correlation of air conductivity (k_a) with saturated hydraulic conductivity (k_s) on the highest lime rate (Lime1.5) showed that both the narrow and the wide coarse pores contribute to gas and water flow on the sandy soil (study site Neubrandenburg: Fig. 6-4 and Fig. 6-5). The improved air flow in the topsoil may lead to an increase in root growth. The share of water-saturated pores as a function of matric potential plays a key role in soil stability. Unsaturated soils are complex three-phase systems consisting of solid, liquid, and gaseous phases, and, according to the effective stress equation of Bishop (1960), the pore water pressure controls the mechanical effects of a change in soil stress (Likos, 2014; Nuth and Laloui, 2008). Thus, water menisci, which exert a contracting effect on particles in the soil matrix, trigger an important process of soil structure formation because their concave curvature attracts soil particles towards each other and increases soil stability towards external loading (Fazekas, 2005). The degree of water saturation and the matric potential of the soil determine the available flow paths of the soil solution and thus the accessibility of surfaces. In soils that have become more aggregated following the lime application, the overall accessibility of surfaces is usually reduced, because the pores in the intra-aggregate system are smaller than the pores in the inter-aggregate system, and therefore the soil solution flows mainly between the aggregates, where it accumulates and transfers substances for chemical and biological processes in the soil. In the intra-aggregate pore system, these processes are slowed and reduced. However, during drainage, the inter-aggregate pore system is soon filled with air and no longer available for exchange and transport processes related to the soil solution. Thus, the finer intra-aggregate pores become decisive in unsaturated soils. Herein, pore continuity plays a decisive role. If this continuity is interrupted, the main flow paths, e.g. for cation exchange processes, are cut off (Booltink and Bouma, 1991). Consequently, Ca²⁺ cannot be transferred into the intra-aggregate pore system to cause structural changes there (Hartmann et al., 1998). Presumably, the tillage

between the two sampling times destroyed the newly formed pore system - therefore differences between the limed plots and the control are hardly recognizable for the water retention parameters 24 months after lime application (Fig. 6-1; Fig. 6-2; Fig. 6-3).

6.5.3 Influence of liming on shear strength

The angle of internal friction (φ) and the cohesion (c) of the aggregated bulk soil are defined by inter-aggregate relations (Horn and Dexter, 1989; Kay and Dexter, 1992), and it is obvious that c is higher in limed plots (Table 6-4). This increase in c is supported by an increasing concentration of polyvalent cations (e.g. Ca²⁺) and enhances soil strength for all studied soil textures (Fig. 6-9) similar to Hartge and Ellies (1977). Thus, both the cohesion and the angle of internal friction are indicators of on increased soil strength towards shear stress (Ayers, 1987). Fattet et al. (2011) determined relations between soil cohesion and aggregate stability, and concluded that aggregation includes bonding mechanisms similar to those that strengthen inter-aggregate structures. Further, the different clay mineralogy at the three different study sites plays an essential role – expansive clay minerals such as montmorillonite provide a high cation exchange capacity and respond more rapidly to lime stabilization than the clay minerals kaolinite and nacrite (Bell, 1996). In our results, the soil's shear behavior shows large variation: already in the plots without lime, c varies considerably with time (Table 6-4). At the study site in Barlt, with the highest clay content (45 %) and the dominant clay mineral montmorillonite (Table 6-2), c was higher in the limed plots 12 months after lime application as well as after 24 months after lime application compared to the control. Additionally, φ was highest in limed plots 24 months after lime application (Table 6-4). However, shear strength was lower on limed plots 12 months after lime application and similar for all plots 24 months after lime application at the study site in Barlt. Nevertheless, rearrangement of the particles and aggregate formation have occurred at all three study sites: Aggregates are stabilized by cementing substances after the addition of lime, resulting in a higher c , which is in accordance with the findings of Zhang and Hartge (1990). At the study sites in Neubrandenburg and Schrecksbach, with nacrite and kaolinite as the dominant clay minerals (Table 6-2), c was higher on limed plots, which corresponds with an increased

shear strength (Fig. 6-9). Shear strength was highest on limed plots for the silty texture (in Schrecksbach) already 12 months after lime application and for the sandy and silty texture 24 months after lime application. The documented changes in c and ϕ suggest a beginning improvement of the soil structure in the limed treatments. The addition of lime enhances the flocculation of clay particles, which initially results in a loose card-house structure. Considering the available organic matter in the A horizon, the formation of clay–organic complexes is another essential process in the observed strengthening of aggregates (Husein Malkawi et al., 1999; Wuddivira and Camps-Roach, 2007). However, soil organic matter can improve the cohesion, and thus the shear strength even in sandy soils (Neubrandenburg: Fig. 6-9), because the associated increased water retention at a given matric potential alters the χ -factor of the effective stress equation (Huang et al., 2021). Thus, the menisci forces remain effective at more negative matric potential values and can support the aggregation and its strengthening (Fredlund and Rahardjo, 1993). The large specific surface area due to the small particles is the reason that clays are stabilized as a result of drainage, while in sandy soils the number and size of the remaining water menisci quickly diminish (Hartge et al., 2014). However, even rather sandy soils (study site Neubrandenburg) can gain a higher strength through organo-mineral bonds. Lime indirectly influences these bonds by enhancing microbial activity due to a reduced acidity in the soil (Mkhonza et al., 2020). Aggregates in sandy soils may be stabilized by a network of fungal hyphae and fine roots (Tisdall and Oades, 1982). Furthermore, roots produce and release polysaccharides in form of mucilage into the soil that glue mineral particles together. These processes lead to the formation of micro-aggregates, which make the grain size appear coarser (= pseudosand), as it was also described by Zimmermann and Horn (2020) and Markgraf and Horn (2007). The newly formed (micro-)aggregates still have a low inherent stability (Table 6-4). The increased shear strength in the sandy and silty soil showed that there is a more homogenous mixing of lime into the soil already during application and first incorporation, and thus, a greater stabilization (already after 24 months) than in clayey soils, where the particle surfaces are only reached by the lime constituents after repeated mixing, and consequently a delayed development of the shear strength takes place (Fig. 6-9).

6.5.4 Impacts of liming on aggregate stability

One of the most useful measures of mechanical aggregate stability is the tensile strength (Y), which is a sensitive indicator of soil structure stability (Munkholm and Kay, 2002). Y can be considered as a measure of resistance against soil deformation in the weakest failure zones (Braunack et al., 1979). The aggregates of all three study sites showed very high Y when dry, but deformed relatively easily when moist (Fig. 6-10 and Fig. 6-11). In air-dried aggregates, the tension in the receding water menisci can generate enough force to create much closer interparticle contacts, which results in a very high strength of the aggregates (Dexter et al., 1988). However, results from our study have shown that clay content and lime application have considerable influence on Y of aggregates in arable soils: Y is generally higher for aggregates with higher clay content (45 % > 24 % > 11 %) as well as higher lime application (Fig. 6-10). Further, Y increased less in aggregates at $\psi_m = -30$ kPa compared to air-dried aggregates (Fig. 6-11). As mentioned above, organic matter can increase the mechanical stability of the soil (Zhang and Horn, 2001) and enhance the effects of a lime application, reflected in an increased aggregate stability at the study site in Barlt (Fig. 6-10 and Fig. 6-11) after the application of lime. A change in the interfacial properties induced by liming leads to an increased accumulation of water-soluble organic matter at the interface between the solid and the liquid phase, and here the adhesion forces between the soil particles in the microaggregates increases and supports a stable aggregate structure (Hartge, 1959a, 1959b).

Even though differences in root development within the aggregates were not explicitly quantified in this survey, we did detect a visible higher density of homogeneously distributed fine roots in soil aggregates from limed plots at the study site in Barlt. Stable microaggregates (20-250 μm) are bound together into macroaggregates (> 250 μm) by e.g., fungal hyphae and fine roots. Oades (1984b) proposed a modification to the aggregate hierarchy concept of Tisdall and Oades (1982) by theorizing the formation of microaggregates within macroaggregates as a result of several processes, such as the reorientation of clay particles around deposited organic debris (Jastrow, 1996). The strength of organo-mineral aggregates depends on the surface area of the involved mineral particles (Kaiser and Guggenberger, 2003). The interactions between roots

and mineral particles also affect aggregation via the release of root exudates such as mucilage (Czarnes et al., 2000; Tisdall and Oades, 1982).

6.5.5 Consequences of tillage between the two sampling times

When soils are exposed to mechanical stresses while in a particularly unstable and fragile state (Gebhardt et al., 2009; Kuncoro et al., 2014), this results in a drastic reduction in stability. Soil tillage plays a major role in this context, especially if it is carried out in spring or autumn under wet conditions (soil water content \geq field capacity). Pores collapse and the hydraulic and pneumatic functions are degraded, as the pore continuity decrease (Fig. 6-4). Additionally, the applied mechanical stresses result in increased pore water pressures, which weaken the structure, especially if cyclic stresses and shearing effects are exerted during tillage (Huang et al., 2021). In order to avoid such negative effects, there is a need for a thoroughly strengthened intra-aggregate as well as inter-aggregate pore system, which can counteract such external forces. In our measurements we documented mostly weakening correlations of k_a with k_s after 24 months with tillage between the two sampling times, which underlines the statement that an adjusted and sustainable soil management is of primary importance.

A solution for the conservation of soil and water resources and the protection of soils against destruction is no-till farming, or at least a management with reduced plowing and soil turnover processes, which proves to be an effective management tool for a sustainable agricultural production (Abid and Lal, 2009). Park and Smucker (2005a) demonstrated the effect of internal particle rearrangements on intra-aggregate pore development, which was only effective when aggregates were subjected to less disturbance as shown for no-till systems. No-tillage improves plant available water and accumulation of soil organic matter (Six et al., 2000). If, in addition to reduced tillage, lime is applied and continuously incorporated into the soil over the long term, the pore system will be improved and strengthened, which in turn guarantees continuous gas fluxes and exchange processes in the soil (Ferreira et al., 2019a; Schack-Kirchner, 1998).

6.6 Conclusion

Lime applications on arable land generated a well-connected pore system, with increased hydraulic and air conductivities due to a shift in the pore size distribution from finer pores towards coarser pores, two years in a row. The rearrangement, interlinkage and connection of soil particles improve the mechanical stability of soil aggregates, shear resistance and the stability of the pore system.

Liming reduces the swelling capacity of clay-rich soils and can thus mitigate structural weaknesses of these soils. Therefore, the trafficability of such soils can be improved by liming.

In current discussions about optimal soil structure development and accessibility of surfaces, physicochemical interactions as well as physical stabilization processes must be considered. From an agricultural point of view, water and gas flow into the topsoil are decisive. An improved air conductivity in the soil in combination with increased aggregate stability can help to prevent erosion and silting on arable land. Consequently, the study of soil aggregation and root development influenced by lime treatments on arable soils should be considered in sustainable soil management systems.

Overall, repeated lime applications in successive years with rates that exceed the recommendation of VDLUFA (2000a) can be especially beneficial in clayey soils with a low initial pH, to achieve a permanent, heterogeneous and sustainable effect in soil strength and maintaining of soil functions, and it can also cause a successful recovery of soil structure after tillage events. However, it must be clear that the positive effects of liming on soil strength are wiped out if tillage disrupts newly created pore systems. Finally, an approach to no-till soil cultivation after lime incorporation is recommended.

6.7 References

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7 Impact of lime application on erosive strength and bulk density of aggregates

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submitted to International Agrophysics

7.1 Abstract

A well aggregated and structured soil with high inter-aggregate strength is favorable on arable land, both to withstand mechanical stresses and for optimal plant growth. The application of lime in form of CaCO_3 can support the formation of a stable soil structure. Therefore, we determined the impact of lime application on erosive strength and density of air-dry aggregates from a Haplic Gleysol with a clay content of 45 %. The lime was applied to the soil in the field with two different rates: 36 dt CaO-equivalents ha^{-1} and 54 dt CaO-equivalents ha^{-1} . The results show that liming significantly increased the erosive strength of aggregates. Lower densities were observed which presumably leads to an improved accessibility of the pores and the particle surfaces within the aggregates due to the application of CaCO_3 . Further, differences between amounts of C and N were determined in the aggregate layers.

7.2 Introduction

Aggregated soils are able to withstand stresses induced by mechanical inputs in order to maintain structure stability with a well-connected pore system that permits the transport of water, gas and nutrients. During mechanical load, the soil structure can carry effective stress but if the load applied to the soil exceeds the internal soil strength the structure is disrupted, effective stresses are reduced, and the internal soil strength decreases (Horn and Fleige, 2003). On arable land, stresses induced by agricultural vehicles can affect soil structure, and if such heavy loads are applied to the soil that the aggregates are destroyed by homogenization and kneading, there is only the primary pore system left. A reduced accessibility of intra-aggregate pore spaces and of the particle surfaces occur which may also result in a loss of nutrients. Stable aggregates provide physical protection of occluded organic matter. When macroaggregates are destroyed by tillage, this leads to the mineralization and release

of nutrients that were formerly protected inside the aggregates (John et al., 2005). Consequently, a high aggregate strength is needed for a sustainable physical protection and accumulation of soil nutrients (Horn, 1990; Jastrow, 1996; Six et al., 2000; Tisdall and Oades, 1982).

Mordhorst et al. (2013), Urbanek et al. (2014) and Park and Smucker (2005c) reported the presence of concentric spatial gradients of soil organic carbon within soil aggregates. The utilization of soil aggregate erosion chambers for the separation of unique concentric layers of aggregates was first introduced by Santos et al. (1997). Park and Smucker (2005a) suggested the calculation of erosive strength of single aggregates based on the erosive forces required to remove 1 g of soil within 1 min from the surface of a soil aggregate rotating along the abrasive wall within an erosion chamber. Urbanek et al. (2014) studied about the soil aggregate stability under three different tillage treatments, and found that the tensile strength of soil aggregates correlated with their erosive strength, suggesting that the tensile strength of an aggregate is controlled by the sum of its concentric layer strengths.

Liming is a well-accepted management practice to decrease the acidity in arable soils, but it can also improve soil structure development and stability (Haynes and Naidu, 1998; Holland et al., 2018; Muneer and Oades, 1989a). Lime application increases the concentrations of Ca^{2+} ions in the soil solution, which promotes aggregate formation due to the development of cation bridges, e.g. between organic material and (clay) mineral particles (Bronick and Lal, 2005) and enhances aggregate stability (Ferreira et al., 2019b; Naveed et al., 2014; Wang et al., 2017). Liming supports diffusive transport processes in the soil because it supports the development of loosely bound aggregates with a coarser pore system (Becher, 1991). Diffusive transport processes in soils are the preferred form of transport, especially in the intra-aggregate pore system, allowing transport from the aggregate surface to the aggregate interior (Park and Smucker, 2005b) The formation of organo-mineral complexes is favored by improved accessibility and aggregate stability is further increased over time (Peth et al., 2008; Smucker et al., 2007) which also protects carbon inside aggregates (Wiesmeier et al., 2012).

The aim of this study was to identify the effects of topsoil lime application on the aggregate stability and the distribution of carbon (C) and nitrogen (N) in different aggregate layers by determining erosive strength as well as pH, C- and N-contents in the exterior and the interior layer of aggregates.

7.3 Material and Methods

7.3.1 Site description

The field trial was established on arable land in summer 2016. The experiment included three treatments: a control with no lime (CaCO_3) application (Lime0), lime application according to the soil-specific recommendations by (VDLUFA, 2000a) to achieve an optimized soil pH (Lime1), and soil-specific recommendations x 1.5 (Lime1.5). The site is located in Barlt, in the western part of Schleswig-Holstein, close to the North Sea in Germany. The soil is a Haplic Gleysol according to IUSS Working Group WRB (2014). The topsoil (0-30 cm) is a silty clay with a clay content of 45 % and an organic carbon content of 56 g kg^{-1} . The lime application rates were $36 \text{ dt CaO-equivalents ha}^{-1}$ in the Lime1-treatment and $54 \text{ dt CaO-equivalents ha}^{-1}$ in the Lime1.5-treatment. The field was managed conventionally with annual tillage and mineral nitrogen fertilization (Frank et al., 2020).

7.3.2 Erosive strength

Aggregate erosive strength was determined with the soil aggregate erosion (SAE) chamber method (Santos et al., 1997). Two concentric layers of equal mass were separated by erosion of air-dried aggregates ($\varnothing \sim 16 \text{ mm}$) as described by Urbanek et al. (2014). The removed outer layer and the remaining aggregate core represent exterior and interior properties of the aggregate (Park and Smucker 2005a). Before and after removing the exterior (ext.) layer, the aggregate was weighed and the volume was determined with a Pyknometer (Geopyc 1360, Micromeritics, Unterschleissheim, Germany). This made it possible to determine the density of the exterior (ext.) layer of the aggregate as well as of the aggregate core representing the interior (int.) layer. The exterior layer was removed from the aggregates by rotating them at 400 rpm in erosion chambers ($\varnothing = 24 \text{ mm}$) on a rotary shaker (IKA KS 260 control, Staufen, Germany) with continuous contact to the abrasive chamber walls, until the weight was about 50 %

of the original weight. To determine this, the actual weight of the respective aggregate was repeatedly determined at certain time intervals (not fixed) during the erosion process.

The external and internal erosive strength of the aggregates (E_s) is equivalent to the erosion forces applied on the surface of each soil aggregate layer, which were calculated from the frictional forces applied on the aggregate surface, using the mass of soil removed during a certain time of the applied centrifugal force (Park and Smucker, 2005a) (eq. 7-1)

$$E_s = C_F / \left[\frac{m(t_n) - m(t_{n-1})}{t_n - t_{n-1}} \right] \quad (\text{eq. 7-1})$$

with

E_s = erosive strength of the aggregates (N g/min)

C_F = centrifugal force (g mm/s²)

$m(t_n) - m(t_{n-1})$ = the mass of the soil aggregate (g) at time t_n and t_{n-1} (min.)

The centrifugal force (C_F) applied on the aggregates in the chambers was calculated following (eq. 7-2):

$$C_F = m(t) [R_{SK} + R_{SAE} - R_a(t)] \omega^2 \quad (\text{eq. 7-2})$$

with

C_F = centrifugal force (g mm/s²)

$m(t)$ = mass (g) at time t (min.)

R_{SK} = radius of rotary shaker movement (mm)

R_{SAE} = radius of erosion chamber (mm)

$R_a(t)$ = radius of aggregate (mm), at time t (min.)

ω = angular velocity (radian/sec.)

7.3.3 pH, C and N of aggregate layers

The eroded material from the aggregate layers was used for chemical analysis. The pH values were measured in 0.01 M CaCl₂ solution (1:2.5 soil:CaCl₂) according to DIN

ISO 11260 (2018). The content of total nitrogen and total carbon (mass-%) was determined with a CNS Element-Analyzer (Vario EL III, Elementar, Langenselbold/Germany) according to DIN ISO 13878 (1998) and DIN ISO 10693 (1997).

7.3.4 Statistical Analysis

The statistical analysis was carried out using the statistical software R version 3.5.1 (R Core Team, 2017). Based on graphical residual analysis, pH-values, C- and N-content were assumed to be normally distributed and heteroscedastic due to different lime application rates at the site. Based on these assumptions, an analysis of variances (One-Factor ANOVA) was performed (Snedecor and Cochran, 1996). All values were evaluated comparing the means of the factor levels treatment and plot for each site separately (Tukey-Test). The significance of the different tests was set at an α -Level of 5 %. Box-and-whisker plots (McGill et al., 1978) were chosen for graphical presentation, showing median (mid-line), 25th/75th percentile (lower/upper border of the box), 95th and 5th percentile (whiskers), 1st and 99th percentile (lower/upper line), outliers (dots) and mean (squares).

7.4 Results & Discussion

The erosive strength (E_s) of the aggregates significantly increased from exterior (ext.) to interior (int.) layers ($p < 0.01$) in all treatments (Fig. 7-1). The E_s in both ext. and int. layers of the aggregates significantly ($p < 0.05$) decreased in Lime1 treatment compared to the control aggregates (Lime0). However, E_s increased (not significantly) in the int. layers of the aggregates from the Lime1.5 plots compared to the control plots (Lime0).

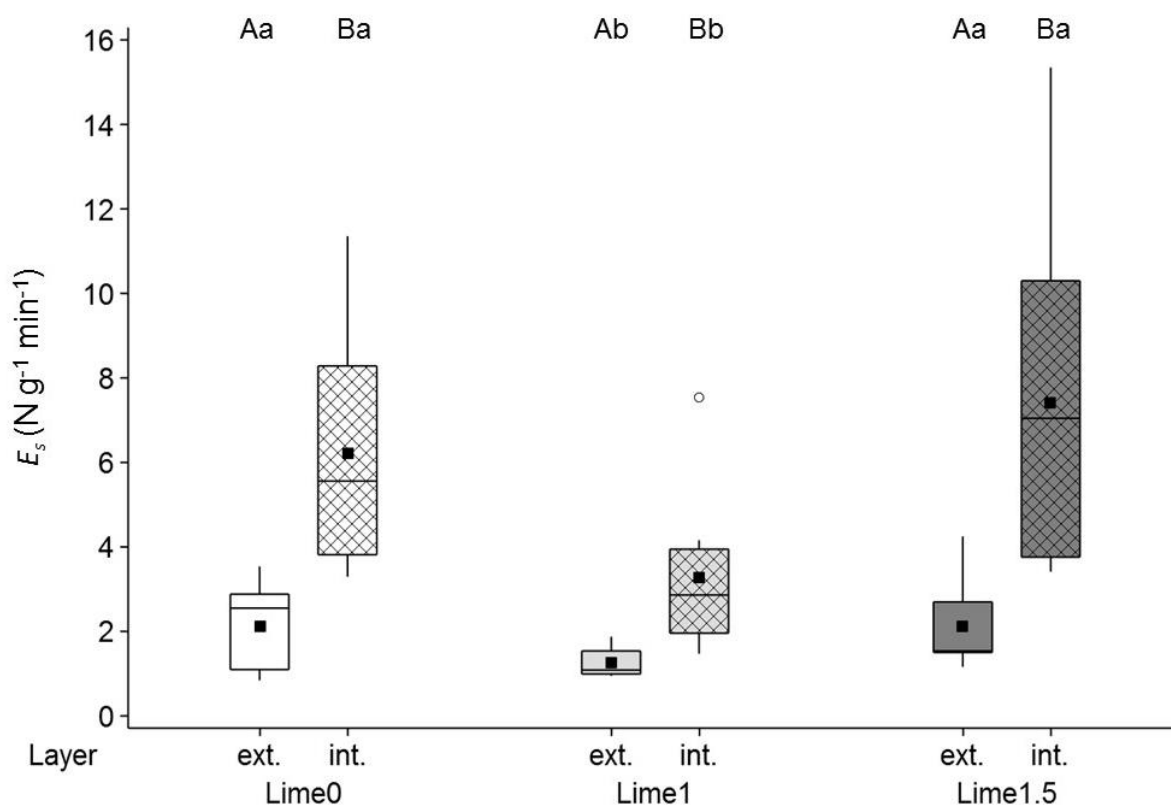


Fig. 7-1: Erosive strength (E_s) of the exterior (ext.) and interior (int.) layers of air-dried aggregates ($\emptyset \sim 16$ mm) obtained from the topsoil of different liming treatments (Lime0: control, Lime1: lime recommendation according to VDLUFA (2000a), Lime1.5: lime recommendation according to VDLUFA (2000a) $\times 1.5$) 32 months after lime application and incorporation ($n = 10$). Squares denote arithmetic means. The capital letters (A, B) identify statistically significant differences between the layers within an aggregate ($p \leq 0.01$). Varying lowercase letters (a, b) identify significant differences among liming treatments ($p < 0.05$).

The densities (ρ) of the ext. layers decreased significantly for the air-dried aggregates from the lime treatments (Lime1 and Lime1.5) compared to the control (Lime0) (Tab. 7-1). In comparison, the densities (ρ) of the int. layers decreased (not significantly) for the air-dried aggregates from the lime treatments (Lime1 and Lime1.5) compared to the control (Lime0).

Table 7-1: Density (ρ) of the exterior (ext.) layer and the interior (int.) with standard derivation (SD) of air-dried aggregates ($\emptyset \sim 16$ mm) obtained from the topsoil of different liming treatments (Lime0: control, Lime1: lime recommendation according to VDLUFA (2000a), Lime1.5: lime recommendation according to VDLUFA (2000a) x 1.5) 32 months after lime application and incorporation (n = 10). *Asterisks indicate treatments significantly different from control at $p < 0.05$.

treatment	ext. layer of aggregates		int. layer of aggregates	
	ρ g cm ⁻³	SD	ρ g cm ⁻³	SD
Lime0	1.64	0.09	1.67	0.07
Lime1	1.41*	0.13	1.64	0.19
Lime1.5	1.38*	0.31	1.62	0.08

Significant changes in both pH values and C and N contents as well as the C:N ratio could not be detected (Table 7-2). However, the pH values in the ext. layers and int. layers were lower in aggregates from the Lime1.5 plots compared to the aggregates from the control (Lime0). The pH values increased in int. layers of air-dried aggregates from Lime1 plots compared to the aggregates from the control (Lime0). The pH value is higher in the int. layer of aggregates compared to the ext. layer of aggregates for the Lime1 treatment. The pH values are lower in the int. layer of aggregates compared to the ext. layer at the control (Lime0) and at the Lime1.5 treatment. Total N-content decreased in both ext. and int. layers of air-dried aggregates from lime treatments compared to the control (Lime0). The content of N is higher in the int. layer of aggregates compared to the ext. layer of aggregates for the Lime1 treatment. Additionally, the content of N is lower in the int. layer of aggregates compared to the ext. layer at the control (Lime0) and at the Lime1.5 treatment. Total C-content decreased in ext. layers of air-dried aggregates from lime treatments (Lime1 and Lime1.5) compared to the control (Lime0) but increased in int. layers of aggregates from the Lime1.5 treatments compared to the control (Lime0). In addition, the content of C is higher in the int. layer of aggregates compared to the ext. layer of aggregates for the Lime1 treatment. In comparison, the content of C is lower in the int. layer of aggregates compared to the

ext. layer at the control (Lime0) and at the Lime1.5 treatment. The C:N ratio is slightly higher in int. layers of all treatments compared to the ext. layers of the aggregates.

Table 7-2: pH (-), total N (mass-%) and total C (mass-%) of the exterior (ext.) layers and the interior (int.) layers of air-dried aggregates ($\varnothing \sim 16$ mm) obtained from the topsoil of different liming treatments (Lime0: control, Lime1: lime recommendation according to VDLUFA (2000a), Lime1.5: lime recommendation according to VDLUFA (2000a) x 1.5) 32 months after lime application and incorporation (n = 2).

treatment	ext. pH	int. pH	ext. N %	int. N %	ext. C %	int. C %	ext C:N	int. C:N
Lime0	6.04	5.98	0.33	0.32	3.48	3.35	10.57	10.59
Lime1	6.05	6.10	0.30	0.31	3.05	3.29	10.25	10.58
Lime1.5	5.93	5.90	0.33	0.31	3.40	3.38	10.45	10.72

The results show that the erosion strength (E_s) of aggregates is not only a suitable parameter for assessing the stability of the pore structure in tillage systems (Jasinska, 2006; Mordhorst, 2013), but also applicable for the evaluation of the structure stabilizing effects of lime application on arable land. Park and Smucker (2005a) already showed that the tensile strength of soil aggregates correlates well with erosion strength and that the tensile strength of the aggregate is controlled by the bonds within the aggregate between the particles, which was also demonstrated in Frank et al. (2021). Park and Smucker (2005a) recognized lowest intra-aggregate porosities in the interior of aggregates from conventionally tilled soils, which has consequences for a low accessibility of soil microorganisms to the organic matter physically protected in soil aggregates. In comparison, we could detect a higher E_s with a simultaneous decrease of the density in int. layers at aggregates of limed plots. The surfaces of aggregates are microbial hot spots (Kuzakov and Blagodatskaya, 2015), and the microbial activity depends on the actual pH in the soil and can be stimulated by a decreasing pH after the addition of lime (Muñoz et al., 2012). Higher E_s of the exterior and interior layers of aggregates as well as the overall increase in aggregate stability might be influenced by repeated swelling and shrinkage processes (Bronswijk and Evers-Vermeer, 1990), which occur preferably in the exterior aggregate layer in the vicinity of already existing

cracks. Repeated shrinkage and swelling causes soil aggregation due to the rearrangement of soil particles from the stress of increasing soil-water suction. Especially the layered silicates of the 2:1 type change with changing water content - they swell and increase their volume. This means that these soils expand when they are wet and contract when they dry out (Kay, 1990). Gradients of microbial activity and root density within the aggregates also influence aggregate stability by secreting extracellular polysaccharides or by fungal hyphae, thereby gluing aggregates together and contributing to increased stability in the soil (Totsche et al., 2018).

Similar to findings of Urbanek et al. (2014), most aggregates in our study presented lower aggregate density in the ext. layers than in the int. layers. Moreover, both the densities of the ext. and int. layers are lowest in aggregates from the limed plots, because of particle rearrangement in a porous 'card-house-structure' during the flocculation and agglomeration processes after lime application, which results in an enhanced intra-aggregate pore accessibility (Park and Smucker, 2005b; Smucker et al., 2007). Due to the short time of our experiment, it is difficult to make long-term statements. The lower aggregate density with simultaneously increased aggregate stability of the aggregates of the limed plots, however, shows that such loosely bound aggregates can result in enhanced cation exchange processes (Hartmann et al., 1998), as well as an increased diffusion of soil solution components from the aggregate surfaces into the aggregate interior (Becher, 1991), where they can enhance the formation of e.g. organo-mineral complexes that further increase aggregate stability (Peth et al., 2008; Smucker et al., 2007). On the other hand, the low erosive strength of aggregate exterior layers indicates the high sensibility of the aggregate exterior against abrasive forces, as they can occur during transport by wind or water erosion. Consequently, organic matter stored in the exterior layers of aggregates is predominantly exposed to mineralization upon exterior aggregate deformation (Mordhorst, 2013). This type of nutrient release can lead to increased mineralization of organic matter, and therefore increased soil fertility, as more nutrients are available in the soil. However, in view of increasing greenhouse gases and ongoing climate change, excessive mineralization of soil organic matter on agricultural land should be avoided. In general, an increasing pH due to the application of lime and the following induced microbial decomposition

are therefore necessary on agricultural land to ensure the mineralization of C and N (Edemeades et al., 1981).

Consequently, liming indirectly improves the transport of nitrogen and the storage of carbon in soils by the rearrangement of particles resulting in increased aggregate strength and soil stability. Our results have shown that higher aggregate stability promotes the development of a stronger structure, which helps to avoid homogenization and kneading under mechanical impact (for example during tillage). Further investigations will be necessary on different textures (sand, loam) and soil types as well as their parent rocks. Additionally, different soil management systems (reduced tillage) should be considered in order to validate over what period the erosion stability will occur and to transfer the results from our field trial to a broader scale.

7.5 Conclusions

- Lower bulk densities were detected on limed plots, which may indicate a reorientation of particles.
- Liming significantly increased the erosive strength of aggregates and may improve the accessibility of pores and particle surfaces within the aggregates

7.6 References

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8 General discussion

In soil science and agricultural practice, it is well-accepted that liming is linked to changes in the soil structure and increases the aggregate stability and porosity of mineral soils. However, physical soil parameters and the influence of lime applications on soil stability and soil structure are not considered in the current advices according to VDLUFA (2000a). The present work intended to analyze whether structural effects of lime applications play an essential role and should be considered in future recommendations. In the context of this thesis conventionally tilled soils were analyzed, which vary in texture, soil organic matter (SOM) and clay content (between 11 % and 45 %). They were subjected to different lime treatments in order to improve physical soil properties like water holding capacity, saturated hydraulic conductivity as well as soil strength, which are all directly linked to soil structure.

8.1 Effects of lime application on chemical and physical parameters

Soil structure is the key for a sustainable agricultural productivity that maintains soil fertility and soil health (Bronick and Lal, 2005). It is already documented that the incorporation of lime on arable land reduces acidity in soils (Chapter 4) and enhances the availability of nutrients for plant uptake (Auler et al., 2017a; Paradelo et al., 2015), but the effects of liming on soil structural changes, as a main factor to be considered concerning lime application recommendations, have not been sufficiently studied on different textures in Germany, until now. Structural changes are observed after application of lime in all texture classes studied for this thesis (Chapters 4-7). Here, lime applied to the soil releases Ca^{2+} ions and carbonates, which promote the rearrangement of clay particles and mineral particles in stable associations and thereby help to develop a porous and stabilized soil structure (Bell, 1996; Metelková et al., 2012). Based on this, the main objective of this thesis was to determine the effectiveness of CaCO_3 application on soil structure stabilization and pore functionality on arable land, in soils with varying clay content. Changes in relevant pore functions, especially for water and gas transport, highly depends on the lime dosage but the developed pore system is often disrupted after tillage. The water retention parameters (plant available water (PAW), air capacity (AC), not plant available water (nPAW)) and

the hydraulic conductivity (k_s) can be classified and assessed according to the Ad-hoc-AG Boden (2005) and DVWK (1995a). However, the effects of liming on these physical properties and, further, influences on structural parameters (shear parameters and pre-compression stress) as well as aggregate properties (tensile and erosive strength) of soils are not well documented, although they directly affect soil chemical processes and indirectly enhance biological activity and plant growth.

The dissolution of lime consumes H^+ ions with an increasing pH in the soil solution. Ca^{2+} and HCO_3^- ions are released and the soil buffer system is based on carbonates again (pH between 8.6 and 6.2). This buffering capacity of the soil is strongly dependent on the type of clay minerals and the amount of soil organic matter, as these factors influence the cation exchange capacity (CEC) (Goulding, 2016). Baldock et al. (1994) and Wuddivira and Camps-Roach (2007) showed that the stabilizing effect of Ca^{2+} is enhanced by simultaneous supply of organic matter. The soil organic matter as well as the soil texture are noted in the recommendations of the VDLUFA (2000a), to reach the target pH ranges (varying between 5.9 and 7.2), but the type of clay mineral is not considered. It is well known that acidic soils can be ameliorated by the application of lime (Goulding, 2016). In the studies of this thesis, liming increased the soil pH in all field trials but the target pH values, as proposed by VDLUFA (2000a), were not reached at all plots (Chapter 4). Especially at the site Struckum the amount of applied lime was not sufficient to reach the recommended pH range according to VDLUFA (2000a), because of restrictions on the maximal annual amounts of lime application in Schleswig-Holstein. At the other study sites, the pH value of both lime treatments decreased again over the years, although the values remained within the recommended range (except for the Lime1.5 treatment in Puch, where the pH decreased under the recommended pH range (6.3-7.0) 20 months after lime application) (Chapters 4 and 5). This can be explained by a reduced accessibility of (clay) particle surfaces for Ca^{2+} due to the presence of other cations on the exchange places.

A further issue is the kind of the pH measurement: the pH analysis defines average values of homogenized samples and does not demonstrate actual in-situ conditions because the accessibility to chemical exchange places is partly hindered due to e.g. soil compaction or aggregation (Hartmann et al., 1998). The pH value of the soil is

subject to fluctuations and changes over time and shows considerable spatial variability within a soil. Under humid climatic conditions, soil acidification occurs gradually due to continuous input or formation of acids. The main sources of acidity are precipitation, respiration of plant roots and soil organisms, microbial degradation of litter and soil organic matter, root uptake of cationic nutrients and oxidation reactions (Bolan et al., 2015; Mkhonza et al., 2020; Zehetner, 2004). Nevertheless, as a consequence of the increase in the soil pH with application of lime in form of CaCO_3 , the dispersive effect of clay particles through the increase of negative charges is minimized with time after the application due to the higher Ca^{2+} and Mg^{2+} concentrations in the soil solution (Frank et al., 2020). In detail, the flocculation or dispersion behavior of soil colloids as well as swelling and shrinkage behavior of soils depend on cation exchange complex, cation valence, and the dominant clay mineralogy (Emerson and Greenland, 1990). The latter and available surface charge can be more important parameters regarding short-term aggregate formation and stability than the absolute clay content (Denef et al., 2002; Lal et al., 1998). The high flocculation capacity of 1:1 minerals and the aggregation by direct linkage of mineral particles are based on the electrostatic attraction between positive edge charges and negative surface charges (Pinheiro-Dick and Schwertmann, 1996). The studied marshland soils in Struckum and Barlt are characterized by the interaction of swelling and shrinkage, promoted by 2:1 clay minerals (Chapter 5).

The swelling of clay minerals is based on their layer structure, their surface charge and attached ions. Two stages of swelling can be distinguished: at the first stage of swelling, cations hydrated in interlayers of smectites or montmorillonites and this process is called intracrystalline swelling (Müller-Vonmoos et al., 1994). The second stage of swelling is called osmotic swelling and is based on the difference between the cation concentration on the mineral surface and the concentration in the pore solution. Cation exchange between the clay particles changes the ion concentration and with it the thickness of the diffuse double layer (Jasmund and Lagaly, 1993). Swelling changes the pore size distribution towards finer pores, so the infiltration of water is no longer assured (Beetham, 2015; Haynes and Naidu, 1998).

It is well known that lime application can limit swelling intensity. There are two ways in which calcium carbonate can directly influence swelling behavior. Firstly, it acts as an

inorganic cement by binding adjacent clay particles together, preventing them from swelling. In an extreme form, this can be an encrustation in which a cluster of clay particles is coated with a layer of calcium carbonate (Rimmer and Greenland, 1976). Induced microbial carbonate precipitation in soil contributes to fill the pore space and bind soil particles together, and thus, this is also a way for cementation in soils (Canakci et al., 2015; Dhimi et al., 2013; Seifan and Berenjian, 2019). Secondly, Ca^{2+} ions compress the diffuse double layer around clay particles while changing the cation concentration (Ulén and Etana, 2014). In general, cations play an important role in soil structure formation and stabilization, as they affect soil particle interactions, for example by strong bonding with Ca^{2+} bridges (Chan and Heenan, 1999). Consequently, lime applications can lastingly increase the concentration of exchangeable Ca^{2+} in the soil solution (Valzano et al., 2001).

In Chapter 5 of this thesis, the elevated pH increased the negative particle charge and thus cation exchange capacity (CEC_{eff}). Further, the percentage of Ca^{2+} in the exchangeable cation fraction increased with increasing amount of applied lime and remained elevated throughout the survey. Additionally, Ca^{2+} ions in the soil solution can reline the surfaces of the clay particles and promote the displacement of dispersive cations such as Na^+ (Bronick and Lal, 2005). High percentages of exchangeable Na^+ weakens the soil structure and are of considerable importance in arid and semi-arid areas and in our studied marshland soils. The dispersing effect of Na^+ ions is based on the monovalent charge acting through a relatively large hydrate shell giving them low adhesive strength (Kuntze, 1965; Prange, 1978). Changes in exchange characteristics can be explained by modifications in the electric potential in the diffuse double layer. Variations in the electric potential modify the preference for either monovalent (Na^+) or divalent (Ca^{2+}) cations: a decrease in electric potential favors preferential adsorption of Na^+ and an increase in electric potential favors that for Ca^{2+} .

Clays, in which external exchange sites dominate and which therefore have low electric potentials (e.g. kaolinite as a non-expandable clay mineral) tend to adsorb Na^+ preferentially (Kopittke et al., 2006). Originally, marshland soils are identified by a high amount of Na^+ . The marshland soils, analyzed in this thesis, were characterized by a high cation exchange capacity with a high Mg^{2+} content on the sorption complex at the study site in Struckum (Chapters 3 and 4), because these soils are desalinated and

decalcified due to the embankment. Ca^{2+} and Mg^{2+} are often considered together in terms of soil structure maintenance, as both cations are known to improve soil structure by bindings between (clay) mineral particle and organic matter. However, the effect of Mg^{2+} on soil structure is less effective than that of Ca^{2+} .

In addition, Mg^{2+} may even have a detrimental effect on aggregate stability compared to Ca^{2+} (Zhang and Norton, 2002). Direct and indirect negative effects of Mg^{2+} can be found in marshland soils, whereby the extent of the effects depends on the clay mineralogy and the electrolyte concentration of the soil solution (Prange, 1978). The direct effect can be described as the adsorption of Mg^{2+} and the resulting deterioration of the soil structure due to the dispersive effect of Mg^{2+} . The latter can be attributed to the higher hydration energy and the larger hydrate shell and a strong interaction between Mg^{2+} and water molecules. The high concentration of Mg^{2+} in the soil solution leads to an expansion of the clay interlayer and the Stern-layer resulting in an increased clay swelling and dispersion (Zhang and Norton, 2002), which reduces soil structural stability e.g. by increasing the vulnerability of soil aggregates to raindrop impacts. The consequence of such dispersion are smaller aggregates, and dispersed particles are potentially able to block pores and promote further surface sealing. This is in agreement with the findings of Dontsova and Norton (2001), who documented that surface sealing is less distinct in Ca-saturated than in Mg-saturated soils (Chapter 5). The indirect effect is based on the affinity of soil with the clay minerals smectite and illite for Na^+ and its adsorption in Mg^{2+} systems. Expansive clay minerals such as montmorillonite, found in the marshland soils studied in this thesis, respond more rapidly to lime stabilization and soil strength than the clay minerals kaolinite and nacrite (Bell, 1996). Montmorillonite has a preference for Ca^{2+} over Mg^{2+} while illite has a preference for Mg^{2+} over Ca^{2+} in the adsorption complex (Rengasamy et al., 2016, 2016; Rengasamy and Marchuk, 2011). Accordingly, a higher proportion of exchangeable Mg^{2+} was found in Struckum with the dominant clay mineral illite. However, the increased proportion of Ca^{2+} in the soil solution after liming reduced the proportion of Mg^{2+} in the cationic exchange complex (Chapter 5).

The results of this thesis also prove that high amounts of fine-grained lime (CaCO_3) can change the Ca^{2+} and Mg^{2+} ratio under humid climate conditions and thus reduce the dispersion in the clayey soils and the related risk of erosion. Wuddivira and Camps-

Roach (2007) explained that the presence of Ca^{2+} and organic matter supports the formation and stability of soil aggregates and results in enhanced physical characteristics (Beetham, 2015; Bell, 1996; Choquette et al., 1987; Hartge and Ellies, 1977) such as an increase in coarser pores and k_s and a decrease of finer pores (Chapters 4 and 5). However, when the soil has a high clay percentage and montmorillonite is the dominant clay mineral, the water-conducting pores clog, resulting in a decreasing k_s (Chapter 5). Consequently, the application of a high dosage (Lime1.5) of fine-grained lime to marshland soils limits swelling and generates a well-connected pore system with increased k_s due to a shift in the pore size distribution from finer pores towards coarser pores and an overall increase in porosity (12 months after lime application). This represents an improved availability and subsequent delivery of water for plants during dry periods.

In the research presented in this thesis, soil structure and the boundary conditions for its formation improved with liming. In detail, the application of lime successfully increased soil pH, cation exchange capacity (CEC_{eff}) (Chapter 5), plant available water (PAW), and soil aeration parameters, and lastingly decreased bulk density (ρ_B) (Chapters 4-6). The relationship between soil structure and lime is long known and several publications have focused on liming effects on soil physical properties as indicators for an improved soil structure (Czeratzki, 1972; Goulding, 2016; Haynes and Naidu, 1998; Holland et al., 2018; Rogasik et al., 2005). Many researchers found a reduced bulk density with lime application in non-tillage systems (Auler et al., 2017a,b). The reduced bulk density is explained by a spatial rearrangement of the pore size. On the contrary, at the study sites included in this thesis, the conventional tillage that was conducted for the incorporation of the lime had a considerable influence on bulk density reduction: the released calcium as well as the carbonates reorganize the loosened soil structure by enhancing flocculation and aggregation of the clay- and mineral particles, which affect the root growth of plants, due to the easier penetration (Anikwe et al., 2016), increasing oxygen availability (Uteau et al., 2013) and oxygen transport functions (Mordhorst et al., 2018), and enhanced k_s (Kirkham et al., 2007) and thus, the availability of water (Horn and Smucker, 2005) (Chapters 5 and 6).

Water flow and availability in the soil is highly influenced by the hydrological cycle which is changing with increasing warming of the atmosphere. These changes can

lead to an increasing number of heavy rainfall events under humid climatic conditions, while the total number of rainfall events tends to decrease (Olsson et al., 2020). Consequently, a stable pore structure in the soil will be even more important to drain water from heavy rainfall events in the future (LAWA, 2017). Extreme climate conditions directly affect the nutrient cycle and plant available water, and thereby indirectly also the soil pH (Reth et al., 2005). The field trials of this thesis were partly exposed to extreme weather conditions during the four trial years. High rainfalls in autumn of the first sampling year (2017) resulted presumably in an increasing H^+ concentration in the soil solution and enhanced leaching of basic cations, which promoted on the one hand soil acidification, and on the other hand, structural collapse (Hartmann et al., 1998) as well as erosion (Finch et al., 2014).

Our results for the saturated hydraulic conductivities (k_s) indicated a more effective water infiltration for all studied texture classes in soil cores from the limed plots (Chapters 4 and 5). Especially in acid soils in the tropics and subtropics, improvements of water infiltration and availability, as well as reduced penetration resistances were found due to lime application (Castro, 1991; Valzano et al., 2001). Similarly to our results, Hoyt (1981) determined also an increased soil water holding capacity in a Luvisol and a Gleysol after lime application, leading to increased rapeseed (*Brassica napus* L.) yields. Lime application induced flocculation of clay particles in the presence of Ca^{2+} (Edwards and Bremner, 1967; Rowley et al., 2018; Yilmaz et al., 2005) and formation of stable macroaggregates (Tisdall and Oades, 1982), resulting in increased k_s . A doubled k_s was observed in a long-term liming experiment in Australia, coincident with increasing total porosity, which was explained by a shift to medium and coarse pores (Kirkham et al., 2007). Findings of Bennett et al. (2014) with an improved aggregate stability and increased hydraulic conductivity of a Red Sodosol in Australia support our results and further indicate an improved pore network with a higher number in preferential flow paths in the lime treatments for all studied textures.

The investigations of soil core samples demonstrated that lime applications did not reduce the soil structure degradation caused by mechanical stress, which was deduced from an overall consistent impairment of water retention parameters as well as high variations in pre-compression stress values (Chapter 4). In addition to texture, degree of aggregation, soil organic matter content and pre-drainage, the

compressibility of the soil also depends on the spatial arrangement of the aggregates and thus the morphology of the pore space, which is determined by the intensity of tillage (Wiermann et al., 2000). Mechanical load creates hydraulic conditions, excess pore water pressure develops in the soil and a change of the effective stress occurs. Water menisci exerting a contracting effect on particles in the soil matrix represent an important process of soil structure formation. The menisci show a concave curvature and pull the soil particles towards each other increasing soil stability towards external stress (Blume et al., 2016b; Fazekas, 2005). When the soil is compressed by mechanical input, the pore space decreases, caused by the destruction of coarser pores, while the finer pores increase (Richard et al., 2001) and the water menisci curvature changes from concave towards convex. The result is a drastic reduction in stability. Soil stability depends not only on the matrix potential but also on the degree of pore water saturation, which is a function of the matrix potential and the pore size (distribution) (Peth et al., 2010). Huang et al. (2021) proved that the pore water pressure increases with loading and decreases with unloading because unsaturated soils are complex three-phase systems consisting of solid, liquid, and gas phases. As water saturation increases, the matrix potential becomes less negative and the χ -factor, described as the “effective stress parameter”, generally varies between 0 and 1 depending on the degree of pore-water saturation. Thus, for a given total stress, the effective stress σ' is reduced, and meniscus force and cohesion are also reduced (Krümmelbein et al., 2013; Peth et al., 2010). The rearrangement of the pores and the change to a porous system induced by the application of lime reduces the pore water pressure during stress in the soils, preventing clay dispersion, kneading and homogenization. Degradation and redistribution of positive pore water pressures are a function of the unsaturated and decreasing saturated hydraulic conductivity, pore continuity as well as hydraulic gradient and are therefore closely related to the texture and (micro-)aggregation of the soils: pore water pressures can be degraded more quickly when flow paths are short and pore continuity is high.

Beside others, the degree of water saturation and the current matric potential of the soil determine the flow paths of the soil solution and thus the accessibility of surfaces at the exchange sites. In soils that are more aggregated after the lime application, the overall accessibility of surfaces is usually reduced, because the pores in the intra-

aggregate system are smaller than the pores in the inter-aggregate system, and therefore the soil solution flows mainly between the aggregates, where it transports and removes substances for chemical and biological processes in the soil. In the intra-aggregate pores system, these processes are slowed down and reduced. During drainage, part of the pore space is filled with air and is no longer available for cation exchange and transport processes related to the soil solution. Fine pores are responsible for the diffusive cation exchange processes and the pore continuity plays a decisive role here; if this continuity is interrupted by tillage, the main flow paths of cations are cut off (Booltink and Bouma, 1991).

In order to get a more detailed insight in the effects of the chemical processes discussed above, both shearing of the structured bulk soils as well as the tensile and the erosive strength of single aggregates underline the intense impact on ecological properties and soil functions.

The effects of lime application on the shear parameters cohesion (c) and angle of internal friction (φ) are explained in Chapters 4 and 6 of this thesis: lime application increased the cohesion between the particles on several study sites with variation in soil texture, similarly to findings of Bohne (1983). As mentioned before, polyvalent cations, such as Ca^{2+} , enhance the aggregation and the formation of soil structure, which changes the pore size distribution and pore continuity. The flocculation and agglomeration of (clay) particles, as induced by liming, causes an increase of intra-aggregate and inter-aggregate porosity. Clay particles are attracted closer to each other and form flocs by the addition of calcium ions, which can be explained by cation exchange between the surfaces of the clay particles (Beetham, 2015). The effect of flocculation was determined in experiments of Beese et al. (1979), who found a strengthening of newly formed aggregates in a loess soil by addition of lime, but during tillage the aggregates were destroyed again. However this homogenization during tillage is less complete, because Aye et al. (2016) could proof that lime application leads to a higher aggregate stability and thereby promotes the development of a stronger structure, which helps to avoid a complete homogenization and the formation of large clods due to smearing and kneading during ploughing. The angle of internal friction (φ) and the cohesion (c) of the bulk soil are determined by inter-aggregate relations (Horn and Dexter, 1989; Kay and Dexter, 1992). Fattet et al. (2011)

determined relations between soil cohesion and aggregate stability and concluded that aggregation includes bonding mechanisms similar to those that strengthen inter-aggregate structures. An increased concentration of polyvalent cations (e.g. Ca^{2+}) in the soil solution promotes the formation of cationic bridges linking clay particles with each other as well as clay particles and soil organic matter (Bronick and Lal, 2005; Muneer and Oades, 1989a). This enhances the formation and stabilization of aggregates, increases c and consequently enhances soil strength (Hartge et al., 2014) to withstand stresses induced during soil tillage. This is one of the essential mechanisms in the soil structure ameliorating effect of liming.

In Chapters 4 and 6 of this thesis, the time-dependency of the soils shear behavior shows large variation: even in the plots without lime, c varies considerably with time. At the study site in Barlt, with montmorillonite as the dominant clay mineral, c increased in the first 32 months and φ at first remained unchanged but decreased in the last months on the plots with lime application. The addition of lime favors the flocculation of the clay particles, which initially results in a loose card-house structure. Husein Malkawi et al. (1999) and Wuddivira and Camps-Roach (2007) stated that organic matter affects the cohesion behavior in a clay–organic complex, forming larger particles, by sharing of ionic forces between clay particles or by bridging of polyvalent cations and organic matter. Secondly, the micro-aggregates formed in this process make the grain size appear coarser (= pseudosand). These bridges of soil organic matter and mineral particles changed the shear parameters, and increased shear strength even in sandy soils (Puch and Neubrandenburg in Chapters 4 and 6). The organic matter can store more water than pure sand and thus increases the menisci forces and the water tension, which in combination with the organic matter leads to aggregation of the mineral particles and further to a change in the internal friction properties (Zhang and Hartge, 1990). Fe (hydr-)oxides and packages of kaolinite, may function as single particles and form a stable aggregate structure with pseudosand textures (Markgraf and Horn, 2007; Zimmermann and Horn, 2020). Fe (hydr-)oxides are found in soils originating from the upper red sandstone (Muggler et al., 1999), which is the parent material for the study site in Schrecksbach (Mader, 1985). The lime-formed micro-aggregates still have a low inherent stability. The increased shear strength in the sandy and silty soil showed that there is a more homogenous mixing of

lime into the soil already during the application and first incorporation, and thus, a greater stabilization (already after 24 months) than in clayey soils, where the particle surfaces are only reached by the lime constituents after repeated mixing, and consequently a delayed development of the shear strength takes place (Chapter 6).

8.2 Accessibility of individual aggregates as affected by liming and aggregation processes

The tensile strength of aggregates (Y) was determined to measure the impact of liming on the mechanical aggregate stability, as shown in Chapter 6 of this thesis. Y is a sensitive indicator of soil structure stability (Munkholm and Kay, 2002) and of the resistance against mechanical soil deformation in the weakest failure zones (Braunack et al., 1979). The Y of the aggregates of the study sites in this thesis increased when dry but decreased when moist. In drying aggregates, the tension in the receding water menisci generates enough force to create increased interparticle contacts, which results in a very high strength of the air-dried aggregates (Dexter et al., 1988). Results from this thesis have shown that clay content and lime application as a function of time have considerable influence on Y in arable soils. Y generally increased with decreasing matric potential (air-dried $> \psi_m = -30$ kPa) and with increasing clay content (study sites Schrecksbach and Barlt) as well as increasing lime application rate as a function of time. The mechanical stabilization of aggregates can be increased by water menisci forces (Fazekas, 2005), clay mineralogy (Barzegar et al., 1995), microbial activity (Tisdall and Oades, 1982) and soil organic matter (SOM) (Zhang, 1994). The study site in Barlt, with the highest SOM content, shows high Y illustrating that organic matter controls aggregate stability and reduces the swelling process in soils (Zaher et al., 2005), wherein especially hydrophobic C-H groups are considered to have a stabilizing effect for soil structure (Abiven et al., 2009) because the organic coating can reduce the wetting of aggregates. Urbanek and Horn (2006) stated that the tensile strength of air-dry aggregates does not depend on the content of organic matter in the aggregates. Nevertheless, the pre-drained ($\psi_m = -30$ kPa) aggregates are influenced by the content of organic matter (Chapter 6), as it was shown for the study site in Barlt. Even though differences in root development within the aggregates were not explicitly quantified in this survey, there were more uniformly distributed fine roots visible in soil

aggregates from limed plots than from control plots at the study site in Barlt (Chapter 6). High clay content increases the shrinkage-swelling capacity and the structural rearrangement during drying and wetting cycles. Particle movement increases the contact areas between the particles and aggregates are additionally stabilized during the process of shrinkage and swelling (Hartge et al., 2014; Horn and Dexter, 1989). During drying, shrinkage occurs and provokes tensile stresses, which results in the development of cracks along planes of weakness (Dexter, 1988). Further, roots support and increase the drying in soils and play a major role in the structural formation in clayey and loamy soils (Bronick and Lal, 2005). One of the most significant plant-induced changes for the soil structure is the formation of continuous macropores by penetrating roots. These macropores support aeration and water movement and storage in the soil (Breemen, 1998). Plant roots prefer to grow in pores (Oades, 1993) and along aggregate surfaces rather than through aggregates (Smucker et al., 2007). Plant and root growth highly modify the structure in the rhizosphere and influences the nutrient and water uptake as well as the gaseous diffusion (Haas and Horn, 2018). The complex interaction which occur in the rhizosphere influences the hydraulic conductivity and the plant root development and can alter soil stability caused by altered physico-chemical properties like surface tension (Haas et al., 2018b).

The pore size distribution of an aggregated soil influences the accessibility of pore surfaces or pore continuity in the long term. The accessibility of the intra-aggregate surfaces is reduced in more structured soils (Horn, 1987, 1990), because the intra-aggregate pore system is always smaller and the pores finer than the inter-aggregate ones (Hartmann et al., 1998). Here, reduced chemical transport processes and access of exchange surfaces are found inside of aggregates with higher bulk density compared to the bulk soil (Jasinska et al., 2006). Consequently, if the concentration of soil particles and the formation of a clay skin at the outer layer of the aggregates reduces the free accessibility of all exchange places, a continuous unbalanced transport between the interior and exterior layers of aggregates is observed (Semmel et al., 1990). In the studies of Ellerbrock and Gerke (2004) and Jasinska (2006) only slight tendencies and not significant results of higher carbon content in the exterior regions have been noticed, probably due to the high heterogeneity among aggregates. It can be concluded that if soil structure remains undisturbed by machinery,

microorganisms and roots as well as preferential flow will occur in secondary pores and the accumulation of organic matter on the aggregate surface will be more pronounced. Moreover, the micro-aggregates protect occluded organic matter against microbial decomposition and leaching, and they influence denitrification and associated N losses (Horn et al., 1994; Lipiec et al., 2009).

The analysis of exterior and interior layers as well as the erosive strength of macroaggregates, as it was presented in Chapter 7, reflects the importance of inter-aggregate pore functions in the evaluation of the influence of liming on aggregate stability. Most aggregates presented lower aggregate density in the exterior layers than for interior layers, similar to findings of Urbanek et al. (2014). The inner and outer aggregate layer density is lowest in the aggregates of the limed plots and can be explained with the particle rearrangement in a porous 'card-house-structure' during the flocculation and agglomeration process after lime application, which also results in an enhanced intra-aggregate pore accessibility (Park and Smucker, 2005b; Smucker et al., 2007). In such less dense aggregates with a coarser pore system, exchange processes are faster (Becher, 1991). This increases the diffusion of mobile soil organic matter components, which are preferentially moved by diffusive transport processes, into the aggregate interior (Park et al., 2007), where the formation of organo-mineral complexes is enhanced and the aggregate stability is further increased over longer times (Peth et al., 2008; Smucker et al., 2007). The non-uniform distributions of nitrogen (N), carbon (C) and pH in the aggregates (Chapter 7) suggest that the formation and functions of macroaggregates are very dynamic processes that influence the distribution of N and C in aggregates (Kavdir and Smucker, 2005; Smucker et al., 2007; Urbanek et al., 2007).

Liming indirectly improves the transport and availability of nitrogen and carbon in soils for roots by directly affecting aggregate and soil stability. Calculated low erosive strength of aggregate exterior regions indicated the high sensibility of aggregate exterior against abrasive forces. Consequently, it might be possible that organic matter that was stored in the exterior layers of aggregates is predominantly exposed to mineralization upon exterior aggregate deformation (Mordhorst, 2013). This type of nutrient release can lead to increased mineralization of organic matter, resulting in leaching and losses of nutrients on the one hand (Goulding, 2000), and resulting in an increased soil fertility, as more nutrients are available in the soil, on the other hand

(Breemen, 1998). Vazquez et al. (2019) stated that the combination of no-till management systems and liming promotes the role of agricultural soils as a C sink by increasing the soil C storage, enhancing microbial activity and increasing soil nutrient contents in the long term. Microbial accessibility is initially limited to the outer parts of the aggregates in no-till systems, but large pores are often of biological origin and represent, for example, root channels with high pore continuity and low tortuosity (Haas et al., 2018a) that connect outer and inner aggregate parts and support a continuous input of fresh roots and microbially produced carbon compounds into the interior of the aggregates (Ananyeva et al., 2013; Kravchenko et al., 2013). In a no-till system, carbon accumulation on aggregate surfaces in the subsoil must be promoted by processes such as solution transport or bioturbation. In general, contents of available C and N, and the microbial biomass are lower in the subsoil as compared to the topsoil (Börjesson et al., 2018). However, tillage helps to displace organic matter into deeper layers, which promotes aggregate stability in the subsoil and aggregate surfaces are important “hot spots” for microbial activity (Kuzyakov and Blagodatskaya, 2015). Ellerbrock and Gerke (2004) stated that the accumulation of organic carbon in exterior layers of aggregates obtained from an arable subsoil were higher compared to aggregate interiors. Consequently, translocation processes of soluble soil organic carbon within the soil profile were attributed to fine root activity and bioturbation, with dissolved organic carbon transport influencing the distribution profile of recently deposited C (Kautz et al., 2013).

N availability for crops increases due to higher N mineralization, but resulting in higher nitrification rates or N losses during periods with no crops on the field (Mkhonza et al., 2020). Soon and Arshad (2005) determined that soil NO_3^- tended to be lower in limed than in unlimed soils during spring and autumn, because of higher N uptake and crop growth in the limed plots. Liming and no-till management enhances the microbial biomass C and N because of an improved soil environment for microorganisms. Increasing N uptake due to N mineralization maximizes soil N availability in an aggregated soil structure (Kavdir and Smucker, 2005) and increases yields on arable land (Soon and Arshad, 2005).

An increased pH value, as it was reached on our study sites (Chapters 4-6), is therefore necessary on agricultural land to ensure the mineralization of C and N (Edemeades et al., 1981; Fuentes et al., 2006), but, in view of increasing greenhouse gases and

ongoing climate change, excessive mineralization (especially under inhibited aeration conditions) of soil organic matter by a loss of physical protection on agricultural land should be avoided. According to VDLUFA (2014), the soil must be supplied with organic matter that meets the requirements of plant growth and environmental protection and, if necessary, the humus balance must be increased with the help of the management method (organic fertilization, crop rotation). A sufficient supply of organic matter is given, if for the respective cultivation system (integrated, ecological) adequate, sustainably high and stable yields are achieved with minimization of nutrient surpluses and losses. Additionally, improving the N efficiency in farming systems, defined as the ratio between N in intended agricultural products and N used to produce them (Godinot et al., 2016), is one of the most important aims in our times. Consequently, there is still a need for measurements during the crop season to observe fluctuations of drying and wetting cycles, root development, changes of gas fluxes and transport process of C and N in the soil and formation as well as transformation of aggregates.

8.3 Effect of soil management and recommendations

Contrary to the positive soil structure formation effect due to liming, soil cultivation counteracts the strengthening effects as derived from the changes of the pore continuity with time. The improved pore connectivity in the limed plots was not rigid enough to withstand the tillage effects (Chapters 4, 5 and 6), but the decline of water retention parameters in limed plots was less intense than in the control plots, especially in soils with a clayey texture, suggesting a higher rigidity of the soil structure towards mechanical disturbance. Soils lose an essential part of the lime induced improvements during the following on-site adjusted tillage management. The kneading of the aggregates during tillage increases the bulk density of the topsoil and the cohesion, combined with a simultaneous loss of inherent stability, which is illustrated by values for the angle of internal friction. A particularly important role in bonding the aggregates or soil particles together have the menisci forces, which can have a stabilizing or destabilizing effect depending on the stress conditions. As a result, if the pore system is not intact, excess pore water cannot be drained off appropriately during loading leading to a mobilization of the soil particles and thus to a reduction in structure (Larson et al., 1989). It could be shown that soil aggregates are vulnerable to disruption by

physical disturbance such as conventional tillage (Bronick and Lal, 2005) and therefore tillage directly negatively affects porosity and pore continuity (index) (Dörner et al., 2012), as it was shown for the study site in Struckum (Chapter 5). Mineralization can occur, which leads to lower amounts of carbon and nitrogen in the soils (Kasper et al., 2009). Furthermore, considering the fact that management practices such as no-tillage are widely used in highly weathered tropical and subtropical soils, these practices in combination with liming for improving physical and chemical soil properties increase the storage of carbon and enhance the water use efficiency for plants (Briedis et al., 2012a; Briedis et al., 2012b; Caires et al., 2006; Garbuio et al., 2011; Joris et al., 2016; Vazquez et al., 2019). Kuhwald et al. (2017; 2020) have shown improved physical soil properties following long-term reduced tillage remain after one-time inversion tillage by mouldboard plough. Consequently, we also need to think about maintaining and strengthening the newly formed aggregates and improving soil structure through liming in combination with conservation (reducing) tillage. Few researchers have focused on alternative forms of tillage in combination with liming in central and northern Europe under humid climate conditions in recent years. Stenberg et al. (2000) determined increased aggregate stability and enhanced microbial activity after reducing tillage depth and lime application in a silty clay loam in Sweden. However, farmers face the problem that on the one hand, the avoidance of tillage impacts is required to meet the time demand to strengthen particle bonds, while on the other hand, lime incorporation by plowing is helpful to obtain an optimal effect concerning the distribution of lime within the soil and to support the interactions of the lime components with particle surfaces. Thus, for future lime application recommendations, the immediate incorporation of the lime directly after application is advisable (Caires et al., 2008; Joris et al., 2016), while the following repeated disturbance of the developing soil structure is known as harmful for soil strength of secondary continuous pores (Peth et al., 2008) as well as the long-term enhanced accessibility of particle surfaces within a finally macroscopically homogenized strong aggregate as the final stage of structure formation.

The presented results have shown that the effects of liming on soil structure stability are superimposed by tillage. It will not be sufficient to rely on existing recommendations, but adapted strategies in sustainable management systems are

necessary to keep soil fertility and thus crop yields stable under humid climatic conditions.

The following recommendations for future lime applications can be made based on the results from this thesis in combination with the discussed literature:

- A general recommendation for farmers should include that lime has to be applied when weather and soil conditions are suitable (according to a sustainable soil management system) - the trafficability of the soils is controlled by the soil moisture.
- Lime should be incorporated in the soil immediately, in order to increase the contact between lime and mineral particles.
- The annual tillage interrupts the process of structural development and even partially reverses it. Consequently, an approach to a reduced soil management after lime incorporation is recommended over several years.
- Higher lime application rates than previously published are recommended for all studied soil textures to generate a well-connected pore system with higher soil strength.
- Repeated lime applications in successive years should be realized in order to achieve a permanent, heterogenous and sustainable effect in the soil and to allow a successful recovery of soil structure after tillage events.

9 Conclusions

The ability of a soil to store and transport water and nutrients for plant production depends to a large extent on the soil structure - i.e. the spatial arrangement of the particles and the voids between them - and its resistance to external stresses. The complex interplay of these mechanisms that determine this resistance, and change under the influence of liming, are summarized in Figure 9-1. In order to characterize soil strength and to be able to assess the soil's ability to store and conduct water, physical-mechanical stability measurements were carried out both on undisturbed soil material and on aggregates. The samples were taken from seven (six of which are presented in this thesis) field trials with three different lime treatments in different geomorphological areas in Germany. From the present results, it can be concluded that lime applications on arable land generated a well-connected pore system with increased hydraulic and air conductivities due to a shift in the pore size distribution from finer pores towards coarser pores. The dispersed clay amount and flocculation values are related to net negative charge of clay particles and further depending on the clay mineral. Thus, especially clay and silt-clay soils are favored by treatments with lime, which are then typically characterized by flocculated particles in a "card-house" arrangement. Clay particles electrostatically attract positively charged ions such as Ca^{2+} , which form bridges holding the particles together. Therefore, these cations can induce flocculation and agglomeration of clay particles. The rearrangement, interlinkage and connection of soil particles improve the mechanical stability of soil aggregates, shear resistance and the stability of the pore system. Consequently, the change to a porous system induced by the application of lime reduces the pore water pressure during stress in the soils, preventing clay dispersion, kneading and homogenization. Soil compressibility is reduced and the soil is less deformable by mechanical loads and existing structures are preserved. Liming reduces the swelling capacity of clay-rich soils and can thus mitigate structural weaknesses of these soils. Therefore, the trafficability of such soils can be improved by liming. In current discussions about optimal soil structure development and accessibility of surfaces, physicochemical interactions as well as physical stabilization processes must be considered. From an agricultural point of view, water and gas flow into the topsoil are decisive. An improved air conductivity in the soil in cooperation with increased

aggregate stability can help to prevent erosion and silting on arable land. Consequently, the study of soil aggregation and root development influenced by lime treatments on arable soils should be considered in sustainable soil management systems. Overall, repeated lime applications in successive years with rates that exceeds the recommendation of VDLUFA (2000a) can be necessary in soils with a low start-pH, to achieve a permanent, heterogenous and sustainable effect and to allow a successful recovery of soil structure after tillage events. The latter disrupts newly created pore systems, consequently, an approach to no-till soil cultivation after lime incorporation is recommended.

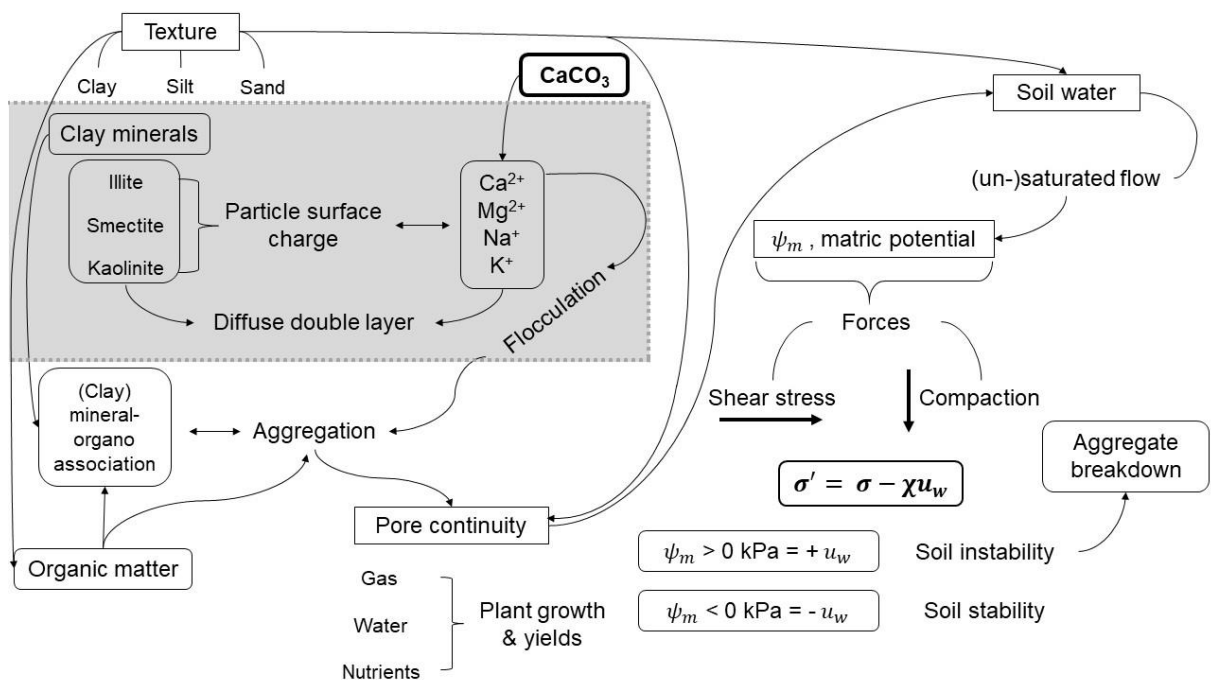


Figure 9-1: Overview of the complex interplay of processes and mechanisms that influence the resistance of soil to external stress and how the influence of added lime (CaCO₃) and the clay minerals present in the soil affect the formation of a stable structure (own figure).

10 References

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