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Soil type, landform and land use as major factors for the storage of organic carbon in agricultural topsoils and subsoils of Bavaria

Stefanie Annelie Sabine Svenja Mayer

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Zusammenfassung

Böden können große Mengen an organischem Material (OM) und damit auch an organischem Kohlenstoff (OC) speichern. Dieser OC ist allerdings nicht gleichmäßig verteilt - weder vertikal im Bodenprofil, noch lateral in der Landschaft. Böden sind die Grundlage für die Produktion von Lebensmitteln, leisten aber auch ihren Beitrag im Klimawandel. Dabei können sie sowohl eine wichtige Senke als auch eine große Quelle für Treibhausgase wie z.B. CO_2 sein. Das Ziel der vorliegenden publikationsbasierten Dissertation war es daher, die maßgeblichen Faktoren für die laterale und vertikale Verteilung des Boden-OC (SOC) zu identifizieren. Besonderes Augenmerk lag dabei auf der Frage, ob die Einflussfaktoren für die OC-Speicherung im Oberboden von denen im Unterboden abweichen. Mit einem Anteil von 47% an der Gesamtfläche ist fast die Hälfte der Fläche Bayerns landwirtschaftlich genutzt. Zusammen mit einer guten Datenverfügbarkeit eignete sich der Freistaat daher gut als Grundlage für die vorliegenden Untersuchungen über landwirtschaftlich genutzte Böden in gemäßigtem Klima. Die Forschungsfragen wurden anhand von drei Studien untersucht, in denen schrittweise das analytische Detail erhöht wurde. So wurden zunächst die OC-Faktoren der Böden in Gesamtbayern (692 Profile), dann innerhalb einer Catena (16 Profile verteilt auf vier Geländepositionen) und schließlich nur innerhalb von Auenböden an zwei verschiedenen Standorten (jeweils vier Profile) in Bayern untersucht. Es wurde zunächst die Gesamtmenge an OC im Boden und in einer Detailstudie die Zugehörigkeit des OC zu bestimmten Fraktionen und dessen Zusammensetzung bestimmt. Neben den Einflussfaktoren sollten auch diejenigen Ober- und Unterböden in Bayern identifiziert werden, die die jeweils höchsten OC-Vorräte besitzen, und es sollte bewertet werden, inwiefern diese von möglichen OC-Verlusten bedroht sind.

Bezüglich der OC Vorräte im Oberboden war die langjährige Landnutzung der relevanteste Einflussfaktor. Oberböden unter konstanter Grünlandnutzung wiesen im Vergleich zu Oberböden unter Wechselland oder unter konstanter Ackernutzung die höchsten OC-Vorräte auf. Nicht nur die Menge des gespeicherten OC, sondern auch dessen chemische Zusammensetzung wurde stark durch die langjährige Landnutzung beeinflusst. Die Zusammensetzung des partikulären organischen Materials der Grünland-Oberböden ähnelte dabei stark derer von nur teilweise abgebauten Pflanzenresten.

Im Unterboden wurden die höchsten OC-Vorräte in alluvialen, kolluvialen und grundwasser-beeinflussten Böden, sowie in tiefliegenden Geländepositionen nachgewiesen. Diese Ergebnisse zeigen, dass Relief und Bodentyp in Bezug auf die Kohlenstoffspeicherung im Unterboden eng miteinander verbunden sind. Das Relief repräsentiert dabei den lateralen Transport von OC-reichem Oberboden aufgrund von Erosion und dessen anschließende Akkumulation in tiefliegenden Geländepositionen. Dort begrenzen die periodische oder permanente Wassersättigung und die damit einhergehende reduzierte Sauerstoffzufuhr den mikrobiellen Abbau von OC. Als Folge dieser Prozesse enthalten alluviale und kolluviale Böden sowie Grundwasserböden hohe OC-Vorräte im Unterboden. Anthropogene Eingriffe wie Bodenbearbeitung verstärken dabei den lateralen Transport und somit die Menge an sedimentiertem OC. Anthropogene Veränderungen des hydrologischen Regimes im Einzugsgebiet durch Flussregulierungen und Drainage können darüber hinaus entweder die Ufererosion verringern (verursacht höhere OC-Vorräte) oder sie verstärken und die Oxidation des Unterbodens hervorrufen (verursacht OC-Verluste).

In den drei zugrundeliegenden Studien wurden die Auen als überaus wichtige OC-Reservoirs identifiziert, da sie typischerweise unter Grünlandnutzung sind (hoher Oberboden-OC-Vorrat) und aus alluvialen Sedimenten entstanden (hoher Unterboden-OC-Vorrat). Die chemische Zusammensetzung des OC im Unterboden der Auen ist dabei sehr variabel. Sie hängt weitgehend von der Zusammensetzung des abgelagerten Sediments sowie vom Grad dessen Wassersättigung ab. In den Unterböden der Auen wurden bemerkenswert hohe Anteile an leichten und vermutlich leicht abbaubaren OC-Fraktionen gefunden - entweder als freie Partikel oder eingeschlossen in Aggregaten. Aufgrund des großen wirtschaftlichen Drucks zu einem intensiveren Anbau, durch Drainage und mögliche Erosionsverluste wurden die großen OC-Reservoirs der Auenböden als gefährdet für OC-Verluste eingestuft.

Die vorliegende Dissertation zeigt, dass die Menge und Zusammensetzung des OC in landwirtschaftlichen Böden in Bayern eng mit dem Bodentyp, dem Relief und der Landnutzung verbunden sind. Die Detailstudien bestätigten dabei die Ergebnisse für Gesamtbayern. Alle drei Studien zeigten den Wert von Auen als hochrelevante OC-Reservoirs (Oberboden und Unterboden). Dabei wurde geschlussfolgert, dass Auenböden potentiell anfällig für die Freisetzung von CO_2 sind, da Landnutzungsänderungen und Drainage die Zersetzung des inhärenten OC der Auen beschleunigen können. Anthropogene Aktivitäten betreffen letztlich nicht nur den OC-Vorrat im Oberboden. Vielmehr beeinflusst das Erbe anthropogener Eingriffe in das Landschaftsökosystem die Menge an OC, die heute im gesamten Bodenprofil gespeichert ist. Dies wird in den alten Kulturlandschaften Europas und Bayerns und dessen Böden besonders deutlich.

Summary

Soils can store large amounts of organic matter (OM) and with that large amounts of organic carbon (OC). This OC is neither evenly distributed in the vertical soil profile nor laterally in the landscape. Soils may be highly productive sites for agricultural use and the production of food. They may also contribute to climate change, as they can be a source of greenhouse gases such as CO_2 – or enhance their mitigation. The aim of this publication-based dissertation was to decipher the factors that led to this heterogeneous vertical and lateral distribution of soil OC. It was further analysed if the factors that control the amount of topsoil OC differ from those of subsoil OC. With a proportion of 47%, almost half of the surface of the Federal State of Bavaria is under agricultural use. Therefore and along with a good data availability, Bavaria was well suited for the studies on agricultural temperate soils. In a stepwise approach of three studies located in Bavaria, the research questions were answered by increasing the analytical detail. Thus, the OC controlling factors were first investigated using a data set on the whole state of Bavaria (692 soil profiles), followed by the analysis of one catena (sixteen soil profiles distributed over four distinct landform positions) and finally of only floodplain soils in two distinct study sites (four profiles, respectively). The analytical detail also increased from broad bulk soil OC quantification to the determination of the specific chemical components of the OM. The objective was further to identify key areas of topsoil and subsoil OC storage and to deduce if these are particularly vulnerable to OC losses.

With respect to topsoil OC storage, the legacy of the land use was the most relevant controlling factor. Topsoils under constant grassland use displayed highest OC stocks compared to topsoils under alternating land uses or constant cropland use. Not only the amount of OC stored but also the chemical composition of this OC was highly affected by the land use. Particulate OM of the grassland topsoils closely resembled the composition of partially degraded plant residues.

In the subsoil, highest OC stocks were found in alluvial, colluvial and groundwater affected soils as well as in low lying landform positions. These findings demonstrated that topography and the generic soil type are closely tied together with respect to subsoil OC storage. The topography represents the lateral transport of OC-rich topsoil due to erosion and its subsequent deposition in low lying landform positions. There, periodical or permanent water saturation and the concomitant reduced oxygen supply limits the microbial decomposition of OC. As a result of these processes, alluvial, colluvial as well as groundwater soils contain high subsoil OC stocks. Human activities such as tillage amplify the lateral OC transport and therefore the amount of buried OC. Anthropogenic modifications of the catchment's hydrologic regime, such as river regulations and drainage, further affect OC storage by altering bank erosion and the oxidation of the subsoil.

The three underlying studies revealed floodplain soils as major OC reservoirs, as most of them are under grassland use (high topsoil OC stocks) and due to their development on alluvial sediments (high subsoil OC stocks). The chemical composition of the subsoil OC in floodplains is highly variable. It is largely depending on the composition of the deposited OC-rich sediments as well as the degree of permanent water saturation. Remarkably high proportions of light and presumably easily decomposable OC fractions were found in the alluvial subsoils – either as free particulate OM or occluded in aggregates. Due to economic pressures to more intensive cultivation as well as drainage and erosional mass losses, it was concluded that the large OC reservoirs of floodplain soils are vulnerable to OC losses.

The presented dissertation demonstrated that the amount and composition of OC of agricultural soils in Bavaria are closely linked to the generic soil type, landform and land use. The in-depth studies confirmed thereby the findings for total Bavaria. All three studies demonstrated the value of floodplain soils as highly relevant OC reservoirs (topsoil and subsoil). Based on the findings, floodplain soils may be prone to the release of CO_2, as land-use changes and drainage may alter the decomposition of the floodplain's inherent OC. To conclude, human activities not solely affect topsoil OC storage. Moreover, the legacy of human activities in the ecosystem of decades and centuries had a strong influence on the amount of OC that is today stored within the whole soil profile. This becomes particularly clear in Europe and Bavaria with its old cultural landscapes and soils.

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Abbreviations

BD	bulk density
DOC	dissolved organic carbon
$F^{14}C$	fraction modern
Fe_{DCB}	dithionite-citrate-bicarbonate soluble iron
Fe_{OX}	acid-ammonium-oxalate soluble iron
fPOM	free particulate organic matter
IC	inorganic carbon
ICP-OES	inductively coupled plasma optical emission spectrometry
MAP	mean annual precipitation
MAT	mean annual temperature
N	nitrogen
NMR	nuclear magnetic resonance
OC	organic carbon
OMF_{fine}/OMF_{coarse}	organo-mineral fraction <20µm/>20µm
$oPOM_{fine}/oPOM_{coarse}$	occluded particulate organic matter <20µm/>20µm
ppm	parts per million
RF	random forest
SOC	soil organic carbon
SOM	soil organic matter
SPT	sodium polytungstate
SSA	specific surface area
TC	total carbon
TWI	topographic wetness index
yBP	years before present (1950)

List of publications and contributions

The presented doctoral dissertation is based on the following three first-authored research articles:

Study I: **Mayer, S.; Kühnel, A.; Burmeister, J.; Kögel-Knabner, I. and M. Wiesmeier (Soil & Tillage Research, under revision): Controlling factors of organic carbon stocks in agricultural topsoils and subsoils of Bavaria.**

Contribution: I conducted the random forest modelling, data evaluation and wrote the manuscript.

Objectives: To identify the controlling factors of topsoil and subsoil OC storage in agricultural soils (cropland and grassland use) at the regional scale of Bavaria and based on these factors, to identify key areas of OC storage within these soils.

Methods: A data set of 692 soil profiles located in Bavaria, Southeast Germany, was used to compute a random forest (RF) model for topsoil (0 – 30 cm) and subsoil (30 – 100 cm) OC storage (kg OC m^{-2}). Thirteen predictor variables (clay content, exposition, inorganic carbon (IC), historical land use, land use, major landform, mean annual precipitation (MAP), mean annual temperature (MAT), pH, rooting depth, sand content, soil type and topographic wetness index (TWI)) were implemented.

Results: The explained variance of the topsoil RF resulted as higher than of the subsoil. Topsoil OC storage was associated with the legacy of land use as highest topsoil stocks were found in sites under constant grassland use. In the subsoil, soil type, landform and TWI resulted as major factors. Highest OC stocks were found in alluvial, colluvial and groundwater affected soils as well as in plains and level lands. Topography (landform and TWI) represented the lateral transport of OC due to soil erosion, which is amplified by human activities. Climatic factors (MAT and MAP) as well as clay content had surprisingly low impact on the model.

Conclusions: The findings demonstrated how closely tied generic soil type, topography, land use and soil OC stocks are. It was concluded that human activities not solely affect topsoil OC storage. Moreover, the legacy of human activities of decades and centuries had a strong influence on the amount of OC that is today stored within the whole soil profile.

Study II: **Mayer, S.; Schwindt, D.; Steffens, M.; Völkel, J. and I. Kögel-Knabner (2018): Drivers of organic carbon allocation in a temperate slope-floodplain catena under agricultural use. Geoderma, 327: p. 63-72.**

Contribution: I conducted the field work and laboratory analysis, carried out data evaluation and wrote the manuscript.

Objectives: To decipher the lateral and vertical distribution of OC stocks along an agriculturally used catena, as determined by the landform position, land use and soil inherent properties.

Methods: Disturbed and undisturbed samples were taken from every horizon of 16 soil profiles located along a catena (backslope, footslope, toeslope, floodplain) in the Bavarian Forest, Southeast Germany. Samples were analysed for total carbon (TC), IC, bulk density (BD), pH, texture, dithionite-citrate-bicarbonate soluble iron (Fe_{OX}), acid-ammonium-oxalate soluble iron (Fe_{DCB}) and ^{14}C content. Soil OC stocks (kg OC m^{-2}) were determined for topsoils (0 - 30 cm) and subsoils (30-120 cm).

Results: Floodplain OC stocks were highest in topsoil and subsoil compared to other landform positions. Also, highest amounts of ^{14}C as well as of Fe_{OX} were found in the floodplain subsoils indicating a young pedogenesis in this landform position. In contrast, stocks on slope positions were lower, subsoil OC contained less ^{14}C and higher amounts of Fe_{DCB}. It was assumed, that the high amounts of young subsoil OC were once deposited as OC-rich sediments in the floodplain by flooding events and that they were hitherto preserved from oxidation due to high water saturation. Dropping of the groundwater table due to drainage left detectable footprints within a few decades in the depth distribution of pedogenic iron oxides.

Conclusions: Geomorphic aspects combined with highly resolved soil data allowed a detailed reconstruction of the pedogenesis and OC dynamics at the study site. The amount of OC stored in the topsoils was mostly determined by land use, whereas subsoil OC stocks highly depended on landform position. The study revealed the major value of floodplains as an OC reservoir compared to other landform positions. The study also demonstrated, that soils may response in relatively short time periods when it comes to changes in the hydrologic system.

Study III: **Mayer, S.; Kölbl, A.; Völkel, J. and I. Kögel-Knabner (2019): Organic matter in temperate cultivated floodplain soils: Light fractions highly contribute to subsoil organic carbon. Geoderma, 337: p. 679-690.**

Contribution: I conducted the field work and laboratory analysis, carried out data evaluation and wrote the manuscript.

Objectives: To deduce the mechanisms of agricultural temperate floodplain soils to stabilise OC by quantifying its allocation to particulate OM and organo-mineral associations and determining its chemical composition.

Methods: Two regulated floodplains under grassland use and of varying parent material in Bavaria, Southeast Germany, were sampled in the topsoil and two subsoil levels. Bulk soil samples were analysed for pH, texture, IC and N. A combined density and size fractionation scheme was applied to obtain six SOM fractions. Chemical properties of the fractions were determined using dry combustion (TC, IC, and OC), solid-state ^{13}C NMR spectroscopy (SOM composition), X-ray diffraction (clay mineralogy) and N_2-BET (SSA).

Results: Despite high inter-site as well as intra-site heterogeneity, remarkably high contributions of light fraction OM (particularly $\mathrm{oPOM}_{\mathrm{fine}}$) were found in topsoils as well as the subsoils of both floodplains. The contribution of organo-mineral associations ($\mathrm{OMF}_{\mathrm{fine}}$) was even higher but only in topsoils. Light fractions were strongly decomposed and lipid-rich in less-aerated subsoils. In well aerated soils with high bioturbation, $\mathrm{oPOM}_{\mathrm{fine}}$ was only weakly decomposed. Charred C was detected at both sites and all depths in the $\mathrm{oPOM}_{\mathrm{fine}}$ composition. Mineral-bound OC storage was higher in calcareous soils compared to decarbonised acidic soils, which was most probably promoted by polyvalent cation bridges.

Conclusions: Contrasting POM compositions are due to the variable composition of OM deposited and buried with alluvial sediments in the floodplain soil (unprocessed plant residues, charred C). It is further modulated by the specific site conditions in the floodplain soil (oxic, anoxic) and turbations of living soil biota. The study illustrates that regulated temperate floodplain soils under grassland use may contain high amounts of OC susceptible to decomposition (such as occluded carbohydrates) throughout the whole soil profile. River regulation and grassland use may promote this OC storage, but increased flooding and associated river bank erosion could cause land losses and the oxidation of OC depending on its chemical composition.

1. Introduction

With an estimated amount of roughly 1,500 Pg, the soils of the world are a major reservoir for organic carbon (OC), which even exceeds the amount of OC stored in the biosphere (560 Pg) and the atmosphere (867 Pg) (Batjes, 2014; Lal, 2018). This reservoir is not static but one compartment of the global carbon cycle that may record gains and losses. With respect to the scientific discourse on global climate change, the question is discussed if soils are a sink or rather a source of greenhouse gasses such as CO_2. Within the last few years, numerous papers have examined the potential of soils to sequester CO_2 and to mitigate greenhouse gases (Conant et al., 2011; Oertel et al., 2016; Paustian et al., 2016; Schmidt et al., 2011; Smith et al., 2018; Smith, 2012). Here, particularly agricultural soils take a central role, as practices such as tillage generally increase the decomposition of SOM and therefore increases the emission, whereas the use of cover crops, retention of residues and organic amendments have the potential to sequester C (Burney et al., 2010; Paustian et al., 2016; Smith et al., 2008; Tubiello et al., 2015). The debate on the sequestration potential of agricultural soils was furthermore intensified after the “4 per mill initiative” launched at the United Nations Climate Change Conference in Paris 2015. As reviewed by Minasny et al. (2017), the overall goal to increase global OC stocks in agricultural soils by 4 per mill to compensate anthropogenic greenhouse gas emissions could be achieved under best management practices. However, not all regions and not all soils are equally suited (Minasny et al., 2017). With 51% of the area of Germany and 47% of Bavaria, agricultural soils constitute a significant proportion of the surface (Bayerisches Landesamt für Statistik, 2016; Bundesministerium für Umwelt, 2016). Hence, agricultural soils hold a fundamental civic value beyond greenhouse gas mitigation due to their function as productive site for food.

The soil OC is not homogeneously distributed and this includes the vertical distribution in the soil profile. In most soils, the amount of OC decreases with depth and largest proportions of the stocks are stored within the uppermost centimetres of the profile (Jobbágy and Jackson, 2000). Looking beyond the topsoils into greater depth revealed that large amounts of OC are stored in the deep soil (Gregory et al., 2014; Jobbágy and Jackson, 2000; Rumpel and Kögel-Knabner, 2010; Rumpel et al., 2012). This is particularly relevant for soils that have developed on alluvial or colluvial sediments. The stratification of allochthonous sediments, i.e. fluvial deposits or material that has moved down a slope by gravitational action as a result of erosional wash, may lead to large accumulations of OC buried in depth (Aldana Jague et al., 2016; Chaopricha and Marín-Spiotta, 2014; D'Elia et al., 2017). The transport and deposition of sediments not only lead to a vertical reallocation but also to lateral fluxes of OC within the field, toposequence or catchment (Doetterl et al., 2012a; Gregorich et al., 1998; Kirkels et al., 2014; VandenBygaart et al., 2012). Studies that follow the catena concept clearly illustrate the variations in OC distribution that result from reallocations of matter depending on the specific landform position. Amongst others, Ritchie et al. (2007) showed that eroding areas contained significantly less OC than depositional sites and that soil OC decreased with increasing slope gradient. The overall amount of such OC reallocations can be enormous, as Doetterl et al. (2012b) calculated a global flux of 403.5 ± 201.8 Tg OC per year due to agricultural erosion.

Besides alluvial and colluvial OC reallocations, not all soils store the same amount of OC. There are several biotic, abiotic, climatic or topographic factors that are assumed to impact OC storage. Wiesmeier et al. (2019) reviewed that climate, topography, parent material, land use as well as biotic factors such as microorganisms, soil fauna, natural vegetation or soil inherent factors such as aggregation, texture, clay mineralogy, specific surface area, metal oxides, Ca and Mg cations affect OC storage - with varying importance depending on the observed spatial scale. As such, metal oxides and specific surface area may be highly relevant for OC storage at the microscale, but the type of land use or climatic factors may superimpose these factors at the regional and global scale (Wiesmeier et al., 2019). Some studies provide evidence that the impact of OC factors may vary depending on soil depth. Based on several global soil data bases, Jobbágy and Jackson (2000) showed that climate and vegetation largely controlled the OC content of the uppermost 20 cm of the soil profile, whereas the clay content as binding agent for SOM, dominated OC content of the subsoil. The increasing amount of available soil data provides an ever more accurate picture of the complex interactions between soil OC factors. However, it is difficult to detect and understand all these interactions between predictors in large data sets. Statistical methods such as data mining and particularly random forest (RF) modelling have therefore become useful tools to analyse these large data sets. The RF model approach as established by Breiman (2001), can be used for variable selection and prediction of the target variable for large data sets. This technique combines the predictions of a large number of decision trees and works with continuous as well as categorical predictor variables (Cutler et al., 2007; Strobl et al., 2008). Results of studies that have implemented RF techniques to determine factors controlling OC storage with respect to soil depth underline the hypothesis that the importance of factors may vary vertically (Hobley et al., 2015; Vos et al., 2019).

For the evaluation if OC is particularly vulnerable for decomposition, it is not only decisive to know the vertical and lateral distribution of OC stocks in the landscape. It is moreover crucial to be aware of the composition and mechanism of stabilisation of the detected OC. Soil OM is not a homogeneous mass, but a very diverse mixture of organic components such as polysaccharides, lignin, lipids, polyphenols, cutin and suberin, originating from plant litter or microbial sources (Kögel-Knabner, 2002). Though, the chemical composition of the SOM may not solely determine the persistence of SOC in the carbon cycle. As described by Tisdall and Oades (1982) the structure of the soil is built up by different sized aggregates of SOM and abiotic particles that are tied by binding agents such as roots or fungal hyphae. These aggregates may also contain SOM that is sorbed to mineral surfaces (Kleber et al., 2007; von Lützow et al., 2006). The specific mechanisms to stabilise SOM are systematically summarised as i) the selective preservation due to the relative accumulation of recalcitrant molecules, ii) spatial occlusion in aggregates and iii) organo-mineral associations (Sollins et al., 1996; von Lützow et al., 2006). As a result, SOM components such as carbohydrates, which are generally supposed to be easily decomposable by heterotrophic microorganisms, may persist for relatively long time in the soil if they are occluded in aggregates and therefore not accessible for microbes. OC bound to mineral surfaces may persist for thousands of years, as indicated by radiocarbon measurements (Rumpel et al., 2002; Torn et al., 1997).

To separate homogenous SOM pools according to their mean residence time in the soil system, a large number of methods have been developed to operationally isolate different SOM fractions. Such fractionation methods may include the physical fractionation of the sample by ultrasonic dispersion, density separation and sieving to specific particle sizes (von Lützow et al., 2007). The fractions isolated by dense liquids such as sodium polytungstate ($3Na_2WO_4 \cdot 9WO_3 \cdot H_2O$) are often denominated as *light* and *heavy* fractions, because the liquid separates those components that are bound to mineral surfaces and have therefore a heavy weight. Ultrasound is applied to isolate SOM that is occluded in aggregates. These fractions are denominated as *occluded particulate organic matter* (oPOM) in contrast to *free particulate organic matter* (fPOM), which is loosely available in the soil matrix. Additional sieving of the fraction is performed as the size of POM mirrors variations in its chemical composition. Hence, smaller particles are stronger decomposed that larger ones (Guggenberger et al., 1995). As several stabilisation mechanisms may act simultaneously, e.g. aggregation and selective preservation or organo-mineral association and selective preservation, the density fractionation is often followed by further chemical analysis of the fractions. Since the 1980'ies, solid-state ^{13}C nuclear magnetic resonance spectroscopy with cross polarization magic angle spinning (^{13}C NMR CPMAS) is increasingly applied to investigate the chemical composition of SOC (Kögel-Knabner and Rumpel, 2018; Preston, 2001) and has become an established analytical method (Kögel-Knabner, 1997).

The presented dissertation examines not only the vertical and lateral distribution of OC stocks in agricultural soils of Bavaria, but also analyses the factors that led to this distribution. It further assesses the vulnerability of this OC towards potential losses. The impetus for this work was provided by the Bavarian State Ministry of the Environment and Consumer Protection within the joint project "Bayerische Landschaften im Klimawandel – Kohlenstoff- und Stickstoffmobilität in Landschaften im Umbruch auf Basis kolluvialer und alluvialer Prozesse" (TKP01KPB-66832). This project determined in an interdisciplinary approach the OC and N stocks of colluvial and alluvial soils in agricultural sites of Bavaria in order to derive adaption strategies for agricultural management in a changing climate.

1.3 Objectives and approach

The aim of this dissertation was to determine the main controlling factors of OC storage of temperate agricultural soils (grassland versus cropland use). It was hypothesised that the impact of the OC controlling factors such as soil inherent properties, climate, topography and land use varies between topsoil and subsoil. Thus, data was analysed with respect to differing soil depths. The objective was further to identify key areas of OC storage within the examined soils. It was then aimed to deduce the soil's mechanisms to stabilise OC, e.g. analyse its allocation to specific SOM fractions and its chemical composition. With respect to the number of soil types and study sites, the variability of the data reduced from Study I to Study III. In contrast, the detail on information within one study site or one specific landform position increased from Study I to Study III. Also, the informational detail on SOM increased from bulk soil data (Study I and Study II) to data on SOM fractions (Study III).

Study I, "Controlling factors of organic carbon stocks in agricultural topsoils and subsoils of Bavaria", was performed in order to identify the relevant controlling factors of OC stocks in agricultural soils (cropland and grassland use) for the federal state of Bavaria. Bavaria was chosen as an example for highly productive soils with a significant proportion of agriculturally used land in a temperate climate in Central Europe. The availability of highly resolved environmental data in Bavaria, e.g. data on soil parameters, climate and topography, guaranteed a reliable data base for the approach. As such, the approach included:

i.) the prediction of OC stocks by computing a random forest model based on a large data set of 692 soil profiles and 13 predictor variables including soil type, topographical factors, climate and information on land use;
ii.) the discrimination of distinct factors for OC storage among topsoils (0 – 30 cm profile depth) and subsoils (30 – 100 cm);
iii.) the identification of key areas for OC storage in Bavaria.

Study I revealed that the factors landform position and land use (cropland vs. grassland) highly affected SOC storage. Therefore Study II, "Drivers of organic carbon allocation in a temperate slope-floodplain catena under agricultural use", was performed in order to closer examine the relationship between soil OC stocks, soil inherent properties and these two factors. The approach combined geomorphic aspects with highly detailed soil data based on 16 soil profiles along a catena in the Bavarian Forest. The study site was representative, as it was agriculturally used for a long period (available historical maps dated back to 1864, settlements in the municipally since at least the 10th century) and the river was regulated. As such, the approach included:

i.) the determination of the lateral and vertical distribution of OC stocks. It was thereby differentiated between topsoil (0 – 30 cm profile depth) and subsoil (30 – 120 cm) and four distinct landform positions (backslope, footslope, toeslope and floodplain);

ii.) the evaluation of the persistence of OC in the diverse landform positions by analysing the proportion of ^{14}C content of the bulk soil OC;

iii.) the analysis of pedogenic oxides as a proxy for the redox conditions as well as the soil development.

Study III, “Organic matter in temperate cultivated floodplain soils: Light fractions highly contribute to subsoil organic carbon”, focussed on floodplain soils, which were identified as key areas of OC storage in Study I and Study II. The study was performed in order to determine the quantity and quality of SOM allocated in representative Bavarian floodplain soils. The selected study sites were representative, as the rivers were regulated, the floodplains were under grassland use and the parent material varied between carbonaceous and carbonate-free. The informational detail on SOM was further increased in Study III, as the allocation of the floodplain OM to characteristic pools differing in stabilisation mechanisms was analysed. As such, the approach included:

i.) the assignment of the floodplain OM to particulate OM and organo-mineral associations by size and density fractionating (six isolated fractions in total) topsoil and subsoil samples (three depth levels in total). The fractionation was followed by the analysis of their OC contents and chemical composition;

iii.) the evaluation of possible OC losses originating from Bavarian floodplain soils based on the identified main OC stabilisation mechanisms.

With the selected approach of increasing the analytical detail from regional data over a catena to a single landform position as well as from broad bulk soil OC quantification to the determination of the specific chemical components of the OM, it was expected to gain further insights into the relevant processes that determine OC storage in agricultural soils.

2. Materials and methods

2.1 Study area and sampling

All three studies were located in Bavaria, Southeast Germany. The federal state of Bavaria stretches over an area of 70,550 km^2 of which 47% are agriculturally used, 37% are under forest and 12% is settlement area and infrastructure (Bayerisches Landesamt für Statistik, 2016). As described in Doppler et al. (2004), Bavaria can be divided into four major geological structures: the Alps in the south with the northward adjacent molasse basin, Mesozoic sedimentary rocks of the Franconian cuesta region in the northwest, crystalline basement complexes of the Bohemian massif in the northeast and the Spessart as an isolated crystalline area in the northwest. All structures may be covered by Quaternary sediments. The climate of Bavaria is classified as warm-temperate, fully humid with warm summer temperatures (Cfb climate, Köppen-Geiger classification) in the transition between maritime in the west and a continental climate in the east (Enders, 1996; Köppen, 1936; Kottek et al., 2006). On average, Bavaria has a mean annual temperature of 7.8° C and a mean annual precipitation of 933 mm (long-term mean 1971 - 2000) (Bayerisches Landesamt für Umwelt, 2018).

Study I was based on a soil survey commissioned by the Bavarian Environmental Agency (LfU) with soil data recorded in an 8 x 8 km grid over the whole federal state of Bavaria (total of 1461 data points). For the presented Study I, the data set was filtered for only mineral soils (no bogs or peat) under cropland or grassland use. This resulted in a total number of 692 soil profiles entering the data analysis (Figure 1).

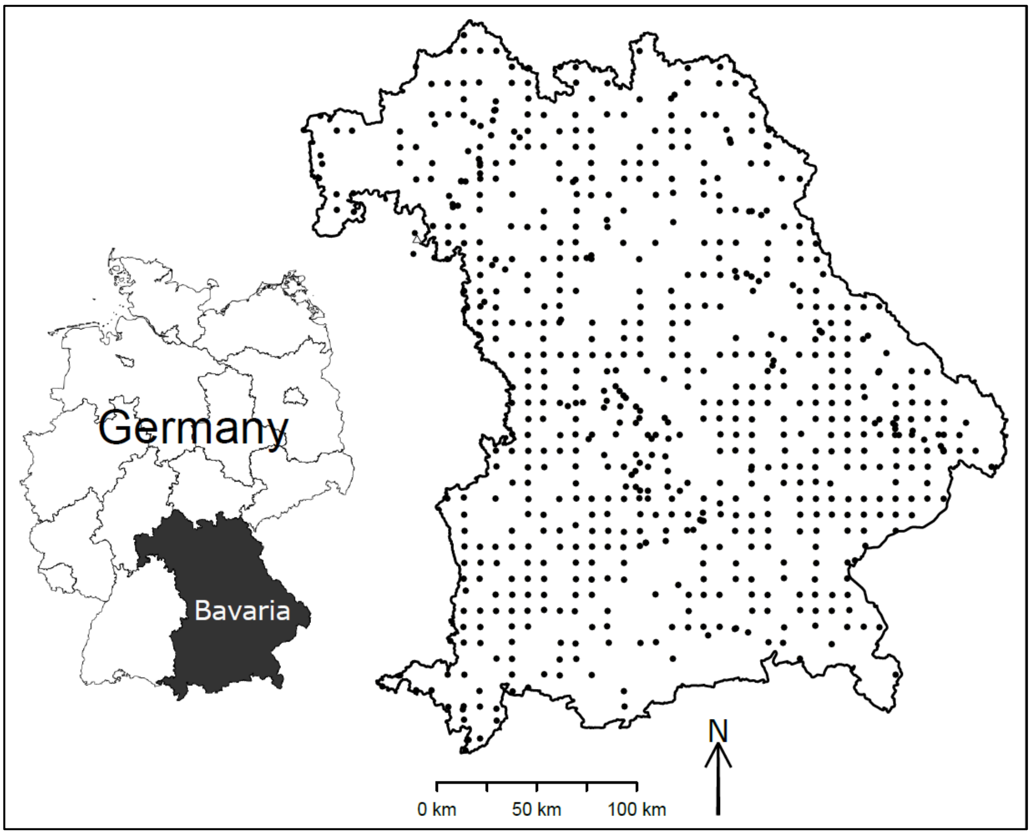

Figure 1: Distribution of sampling points of Study I (n = 692).

For Study II, the site was located in the low mountain ranges of the Bavarian Forest, east of the city of Regensburg (49°02'06"N, 12°15'10"E) (Figure 2). There, at the lower reaches of the Otterbach Valley, 16 soil profiles were excavated in a transect from the eastern slope of the Otterbach River over the floodplain to the western slope (Figure 2). According to the dominant geomorphic processes and deposition dynamics, soils were assigned to four distinct landform positions within this catena: backslope (soil profiles OBS1 – OBS4), footslope (OFS1 – OFS3), toeslope (OTS1 – OTS3) and floodplain (OFP1 – OFP6). All slope positions were constantly used as cropland, whereas floodplain positions were under constant grassland use for more than one century. In the lowlands of the study site, artificial drainage tiles and ditches were installed. Weirs regulated the river. Based on the interpretation of historical maps, river straightening by cutting up meanders was carried out between 1864 and 1951, and the riverbed was cleaned during land consolidation works, which finished in 1992. The bank crest of the river lay approximately 100 cm above the channel (Figure 3), which may have been caused by riverbed incisions after the regulations. The parent material of the soils consisted of weathered granite of the Bohemian massif and solifluidal deposits with substantial contributions of decarbonised quaternary loess (Völkel, 1995). With a mean annual temperature of 9.2° C and a mean annual precipitation of 981 mm (long-term mean 1981 – 2010) (Deutscher Wetterdienst, 2017), study site II is warmer and slightly more humid than the average of Bavaria.

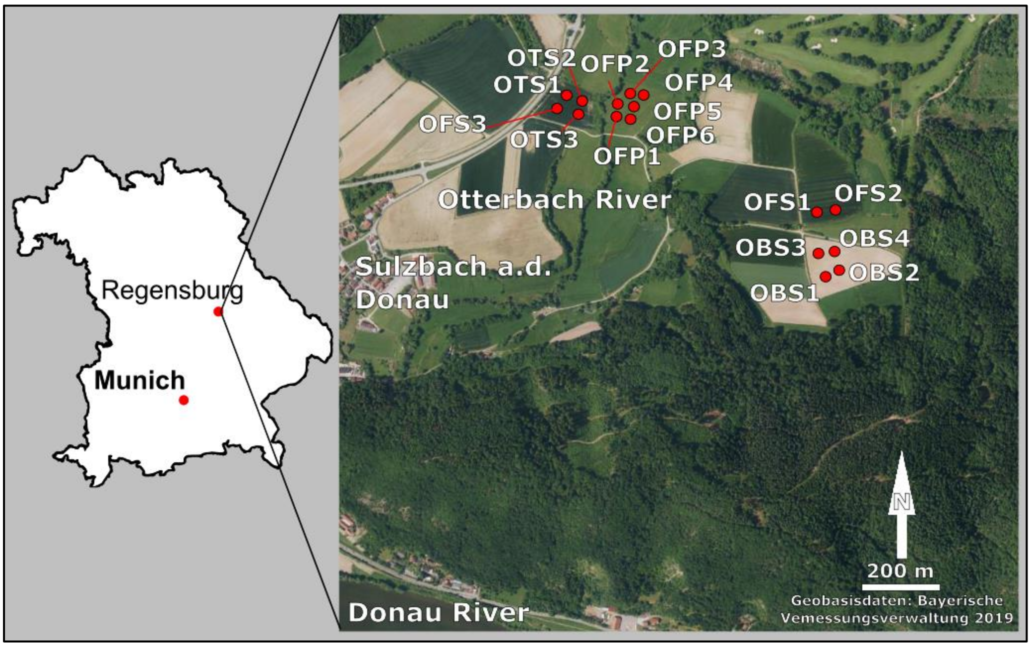

Figure 2: Map of the study area of Study II including the location of sampling points.

Figure 3: Soil profile at the eroded bank crest of the Otterbach River (profile data not shown) (Foto: S. Kriegs, 2015).

Focussing on floodplain soils in Study III, additionally to soil data of the floodplain of Study II, four soil profiles (AFP1, AFP2, AFP3, AFP4) were excavated in a floodplain of the Ammer River located in the municipality of Rottenbuch in the Alpine Foreland (47°44'13" N, 10°58'22" E) (Figure 4). In contrast to the Otterbach site, no artificial drainage measures were observed in the floodplain. Weirs regulated the river. The parent material of the soil was a broad mixture of deposits of moraine till (Würm glaciation) and layers of the tertiary Lower Freshwater Molasse, which have been incised by the Ammer River. It also contained deposited sediments and bed load of Rhenodanubian Flysch, Hauptdolomit and Dachstein limestone originating from the Northern Calcareous Alps (Bayerisches Geologisches Landesamt, 1996). The floodplain was used as extensive grassland. The mean annual temperature was 7.2° C and mean annual precipitation was 1,175 mm (long-term mean 1981 – 2010) (Deutscher Wetterdienst, 2017), so study site III is about as warm but more humid than the Bavarian average.

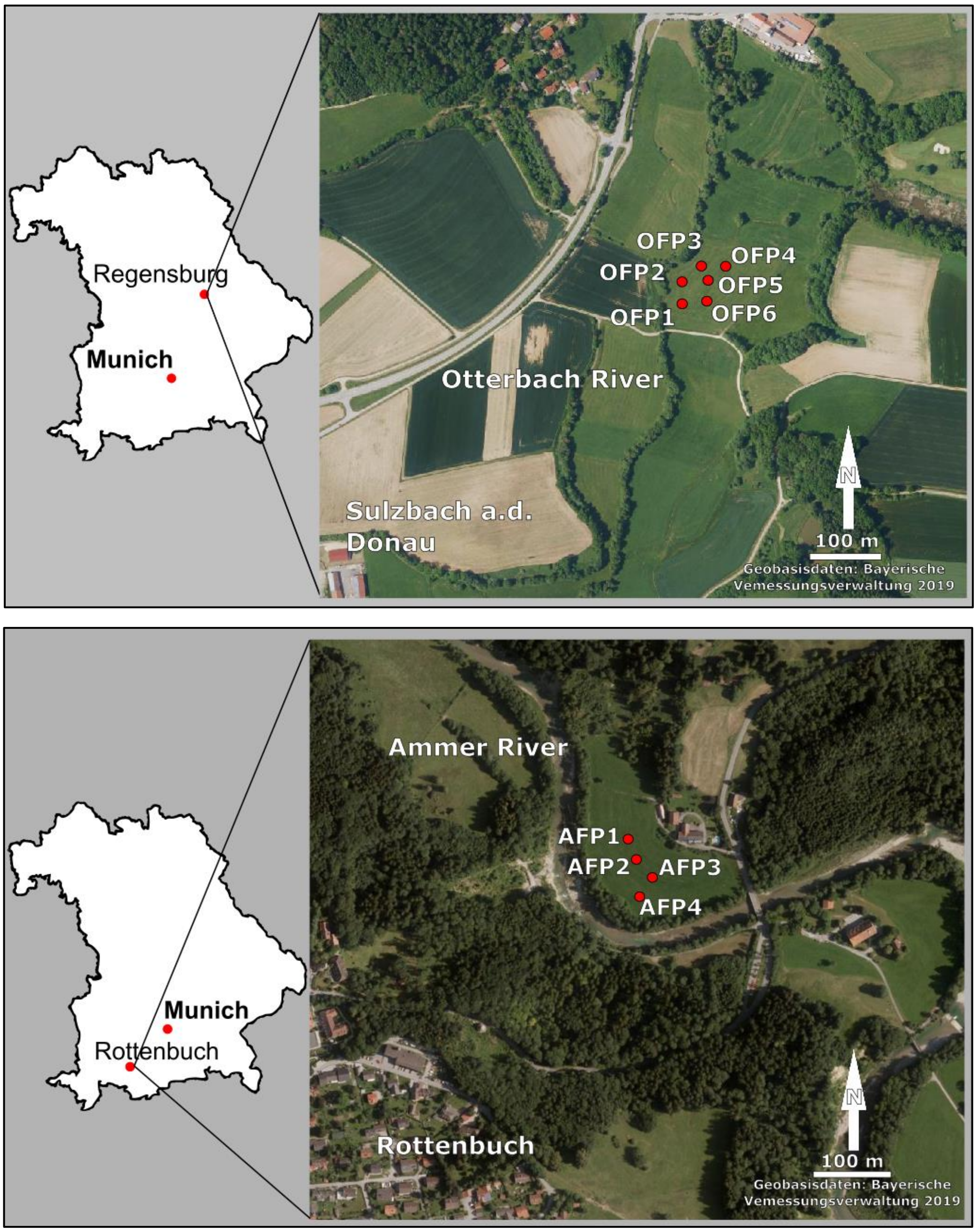

Figure 4: Maps of the study areas of Study III including the locations of sampling points.

Soils of all studies were classified according to the German soil classification (KA5) (Ad-hoc-Arbeitsgruppe Boden, 2005). For Study I, soil classes of the KA5 were translated to equivalent reference soil groups according to the World Reference Base (WRB) for soil resources (IUSS Working Group WRB, 2015). Soils of Study II and III were also classified according to the WRB.

Samples of all studies were taken by horizon including disturbed, as well as undisturbed samples for BD determination. In Study I, soils were sampled down to a depth of 100 cm or at least to the parent material. In the case of Study II and III, samples were taken to a maximum depth of 180 cm depending on operational safety and/or the presence of ground water.

2.2 Analytical methods

Samples of Study II and III were air dried and sieved to <2 mm. Samples of Study I were freshly sieved before drying. Table 1 gives an overview over the determined soil parameters and applied methodologies of the three presented studies.

Table 1: Overview of determined soil parameters and applied methodologies for Study I, II and III.

Parameter	Methodology	Study
BD	Drying at 105°C and weighting of undisturbed samples	I, II, III
Clay mineralogy	X-ray diffraction (D5000 diffractometer, Siemens, München, Germany)	III
Fe_{OX}	Acid-ammonium-oxalate extraction followed by ICP-OES (Vista Pro CCD Simultaneous, Varian, Darmstadt, Germany)	II
Fe_{DCB}	Dithionite-citrate-bicarbonate extraction followed by ICP-OES (Vista Pro CCD Simultaneous, Varian, Darmstadt, Germany)	II
^{14}C	Accelerator mass spectrometry carried out at the CologneAMS centre	II
OC composition	Solid state ^{13}C NMR CPMAS spectroscopy (Brucker DSX 200 NMR spectrometer, Karlsruhe, Germany)	III
IC	Thermal gradient method (RC612, LECO, Saint Joseph, USA)	II, III
IC	Muffling at 500°C followed by dry combustion at 1,000° C	I, III
N	Dry combustion at 1,000° C, Elemental Analyzer (EuroEA, HekaTech, Wegberg, Germany)	II, III
pH	0.01 M $CaCl_2$ solution (1_{soil}:2.5_{liquid}) (pH 340, WTW, Weilheim, Germany)	I, II, III
SSA	N_2 BET gas adsorption-desorption (AUTORSORP-1, Quantachrome, Odelzhausen, Germany)	III
TC	Dry combustion at 1,100° C, Elemental Analyzer (EuroEA, HEKAtech, Wegberg, Germany)	I, II, III
Texture	Sieving and sedimentation (Köhn)	I
Texture	Wet sieving followed by X-ray sedimentation technique (Sedigraph III Plus, Micromeritics, Aachen, Germany)	II, III

2.2.1 OC determination and calculation of OC stocks (Study I, II, III)

All bulk soil samples as well as fractions were analysed for TC as determined by dry combustion at 1,000° C using an Elemental Analyser. For soils located at the Ammer River as well as for Study I, the content of OC was calculated as the difference between TC and IC. Concentrations of IC were assumed as negligible at the Otterbach River, as soil pH resulted in strongly to moderately acidic pH, and tests on individual samples revealed a concentration of 0.3 ± 0.3 mg IC g^{-1}. Therefore, TC was set equal OC for this study site.

Soil OC stocks of all three studies were calculated using the equation:

$$SOC_{h_z} = \sum_{s}^{h_z} OC_s \times h_s \times BD_s \times \left(1 - \frac{SC_s}{100}\right) \qquad (1).$$

The variable SOC_{hz} corresponds to the OC stock of all horizons h of the soil profile z. Factor h_s corresponds to the thickness of the horizon s multiplied with BD_s. Factor SC_S is the mass fraction of the coarse material >2 mm of the horizon s.

To compare soil characteristics of contrasting depths, generic horizons at three depth levels of the profiles investigated in Study II and III, accounting for the topsoil and two subsoil horizons, were chosen for further chemical and physical analysis (^{14}C content, pedogenic Fe, texture, SOM fractions and SSA). The horizon referred to as *top* was the upper-most horizon of every profile. *Sub2* corresponds to horizons located between 38 and 57 cm and *sub2* between 67 and 128 cm profile depth at the Otterbach site. As soil profiles at the Ammer site were shallow, corresponding depths shifted upwards: 24 – 66 cm (*sub1*) and 41 – 90 cm (*sub2*).

2.2.2 Random forest modelling and its data base (Study I)

In Study I, the controlling factors of topsoil (0 – 30 cm) and subsoil (30 – 100 cm) OC stocks were determined using 13 predictor variables: clay content, exposition, historical land use, IC, land use, major landform, MAT, MAP, pH, rooting depth, sand content, soil type and TWI.

The laboratory analyses of the BD, clay and sand content, IC, OC and pH were executed by the Bavarian Environmental Agency (LfU). Data on the historical land use originated from the land taxation data of the Bavarian State Ministry for

Finance (BayLfSt). Between the thirties and sixties of the 20th century, all agricultural sites of Germany were mapped and evaluated in a 50 x 50 m grid. Since then a regular review every 20 years is executed. The predictor variable historical land use includes four classes: sites permanently used as cropland (C), sites permanently used as grassland (Gr) as well as sites with periodical alternations between C and Gr with a dominance of grassland use (GrC) and those with a dominance of cropland use (CGr) (Rösch and Kurandt, 1950). The predictor variable major landform describes the landform by its morphology and is subdivided into four major classes: crest (culmination point of the hill), slope (located between crest and level land), level land (2 - 6° inclination) and plain (<2° inclination). Rooting depth was taken from the 1:1,000,000 map “Soil Depth in Germany” (Bug, 2015). Data on MAP and MAT were recorded by the German Weather Service (DWD) (1 km resolution, long-time mean 1981 - 2010). The TWI was implemented as a proxy for potential soil moisture and the accumulation of depositional sediments (Sørensen et al., 2006). As a result of the specific contributing area (SCA) and its slope α, the TWI was calculated following the equation:

$$TWI = \ln\left(\frac{SCA}{tan\alpha}\right) \qquad (2).$$

The underlying topographic data was based on a Digital Elevation Model (DEM) (Bavarian Surveying and Mapping Authority, BVV) (25 m resolution).

The modelling was computed using the random forest (RF) machine learning technique established by Breiman (2001). With RF, large data sets can be analysed for prediction as well as for variable selection. For computation of the model, the “party” package Vol. 1.3 – 0 developed by Hothorn et al. (2006) was implemented in the software R (R Core Team, 2016). To minimize bias between the individual trees (n = 1,000) of the forest, bootstrap aggregating without replacement of the sample was executed (Strobl et al., 2007). Its performance was evaluated by the coefficient of variance (R^2_{OOB}) as well as the root-mean-squared error ($RMSE_{OOB}$) over the averaged out-of-bag predictions, i.e. the samples that were not included in the bootstrapped samples (Liaw and Wiener, 2002) following the equations:

$$RMSE_{OOB} = \sqrt{\frac{1}{n} \sum_{i=1}^{n} \left(z_i - \hat{z}_i^{OOB}\right)^2} \quad (3),$$

$$R^2_{OOB} = 1 - \frac{MSE_{OOB}}{Variance_z} \quad (4),$$

where z_i corresponds to the measured OC stock of the depth increment *i* and $\hat{z}_i$ is the predicted value over the averaged out-of-bag samples. Most important variables were selected on the base of the variable importance area under the curve (Janitza et al., 2013). The relative variable importance area under the curve was calculated in order to increase comparability between the two models. This is relative to the summed variable importance of the respective RF (Hobley et al., 2015).

2.2.3 ^{14}C content, pedogenic Fe and texture (Study II)

Study II assesses the vertical and lateral OC distribution as well as OC relevant soil properties in a catena. Therefore, not only OC concentrations and stocks were calculated but also ^{14}C content, pedogenic Fe and texture measured. Pedogenic Fe was extracted as dithionite-citrate-bicarbonate soluble fraction (Fe_{DCB}) and acid-ammonium-oxalate soluble fraction (Fe_{OX}) (Holmgren, 1967; Schwertmann, 1964). This was followed by inductively coupled plasma optical emission spectrometry (ICP-OES). The Fe activity ratio (Fe_{OX}:Fe_{DCB}) was calculated as a measure of pedogenesis according to Schwertmann (1964).

The ^{14}C content of selected bulk soil samples was determined by the CologneAMS centre via accelerator mass spectrometry (AMS). Radiocarbon data was reported as Fraction Modern ($F^{14}C$) with corresponding conventional radiocarbon ages (^{14}C age) given as years before 1950 (yBP) (Reimer et al., 2004; Stuiver and Polach, 1977).

For texture analysis, samples were treated with 30%-H_2O_2 to remove OC before ultrasonic dispersion at 450 J m^{-1} in a 0.025 M $Na_4P_2O_7$ solution. Wet sieving was followed by the X-ray sedimentation technique using a sedigraph.

2.2.4 Combined density and particle-size fractionation, ^{13}C NMR spectroscopy and SSA measurements (Study III)

Study III focuses on the quantity and quality of OM in floodplain soils, so the following analysis were applied for soil profiles OFP1, OFP2, OFP4 and OFP6 of the Otterbach site and AFP1, AFP2, AFP3, AFP4 at the Ammer site. They were further applied at the three depth levels *top*, *sub1* and *sub2* in each soil profile. In order to quantify the allocation of SOM to particulate OM and organo-mineral associations, bulk soil samples were analysed following a combined stepwise density and particle-size fractionation procedure. The implemented procedure was elaborated by Kreyling et al. (2013) for the application in dolomite-derived soils. They used distinct densities of a sodium polytungstate solution to further separate mineral associated OM from OM-free heavy particles such as dolomite. To ensure comparability of the results, the procedure was customised for Study III on the basis of preliminary tests and applied to the samples of both Ammer and Otterbach site. An overview over the procedure and the resulting fractions is given in Figure 5. Briefly, sodium polytungstate of a density of 1.8 g cm^{-1} ($SPT_{1.8}$) was added to 30 g bulk soil. Floating free particulate organic matter (fPOM) was separated. The deposited material was dispersed ultrasonically at 450 J ml^{-1} to destroy aggregates and subsequently centrifuged. Now floating particles were associated with formerly aggregate-occluded POM (oPOM) and separated. To separate the lighter organo-mineral fraction (OMF) from OM-free mineral fraction, the remaining material was suspended in sodium polytungstate of a density of 2.4 g cm^{-1} ($SPT_{2.4}$), subsequently centrifuged and decanted. To account for differences in the degree of degradation, the oPOM fraction as well as OMF were wet sieved to 20 µm resulting in four fractions: oPOM <20µm ($oPOM_{fine}$), OMF <20 µm (OMF_{fine}), oPOM >20 µm ($oPOM_{coarse}$) and OMF >20 µm (OMF_{coarse}). All fractions were desalted with deionized water before freeze-drying and weighting.

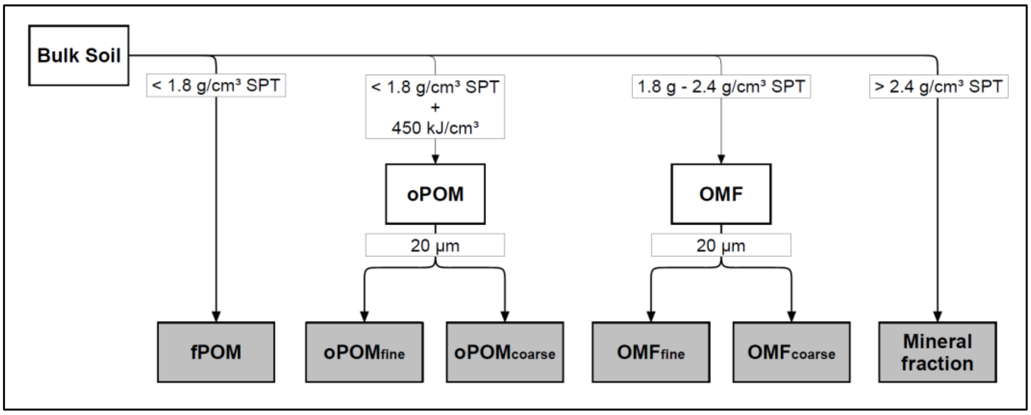

Figure 5: Scheme of the fractionation procedure.

As oPOM$_{fine}$ and OMF$_{fine}$ accounted for up to 80% of total OC, the composition of these two fractions was further characterised using ^{13}C nuclear magnetic resonance (NMR) spectroscopy with cross-polarization magic angle spinning technique (CPMAS). Signal intensities of the chemical shifts relative to the standard tetramethylsilane (= 0 ppm) were integrated according to four spectral regions: alkyl-C (0 – 45 ppm), O-alkyl-C (45 – 110 ppm), aryl-C (110 – 160 ppm) and carboxyl-C (160 – 220 ppm) (Wilson, 1987). The ratio between alkyl:O-alkyl-C was calculated to estimate the degree of decomposition (Baldock et al., 1997). A second integration routine following the molecular mixing model of Baldock et al. (2004) was applied to infer biomolecular structures. These spectral regions were: 0 – 45, 45 – 60, 60 – 95, 95 – 110, 110 – 145, 145 – 165, 165 – 215 ppm.

To evaluate the OC-coverage of the minerals of the OMF$_{fine}$, the specific surface area (SSA) of selected samples of this fraction (OFP1 and AFP2 at *top*, *sub1* and *sub2*) was determined by N_2 gas adsorption-desorption at 77 K and the application of the Brunauer-Emmett-Teller equation (Brunauer et al., 1938). Samples were measured before and after OC removal using 1 M NaOCl at pH 8 (SSA$_{OC}$ and SSA$_{NaOCl}$, respectively). The loading of the mineral surface was calculated as the ratio between OC content of the fraction before OC removal and the SSA$_{NaOCl}$. The OC coverage of the mineral was calculated according to the equation:

$$coverage = \frac{SSA_{OC} - SSA_{NaOCl}}{SSA_{NaOCl}} \qquad (5).$$

2.3 Statistical analysis

All statistical analysis were performed in the software R (R Core Team, 2016). Normality and homoscedasticity of the data was tested using the Shapiro-Wilk and the Levene's test. Welch's ANOVA was used for testing significance in case of unequal variances. The repeated t-test was used in case of dependent samples (Study II). Both were followed by the Scheffé post-hoc test. Due to insufficient observations of soil types (Study I, $n_{Podzols} = 1$), no statistical testing was applied for this grouping variable. The level of significance was chosen at $p < 0.05$.

3. Results and discussion

3.1 Controlling factors of topsoil OC storage

Study I and II promoted the assumption that topsoil (0 – 30 cm profile depth) OC storage is largely controlled by the legacy of land use. With 34% variable importance, the historical land use was the most important predictor variable for topsoil OC stocks in Study I. This resulted in highest amounts of topsoil OC found under grassland use. In Study II, topsoil OC stocks under constant grassland use in the floodplain were significantly higher than stocks under constant cropland use. OC stocks under constant cropland did not statistically differ between the distinct slope positions observed in Study II.

It is widely assumed that grassland use promotes the storage of OC in contrast to cropland use (Guo and Gifford, 2002; Jones and Donnelly, 2004). Grasslands have intensively rooted sods which produce high inputs of fresh OM into the topsoil (Kuzyakov and Domanski, 2000). Study III provided evidence that not only the amount but also the composition the SOM found in the topsoils of grassland floodplains was largely controlled by the grassland use (Baldock and Skjemstad, 2000; Li et al., 2017). The high amounts of topsoil OC were to large parts organo-mineral associated in OMF_{fine}, followed by $oPOM_{fine}$. The ^{13}C NMR spectra of the $oPOM_{fine}$ matched with partially degraded, carbohydrate-rich plant residues, as large proportions of O-alkyl C, followed by alkyl and carboxyl C were found. This is indicative for regular inputs of fresh OC as typical for grassland use. On the other hand, it has been shown that constant cropland use leads to OC losses due to aggregate breakdown by ploughing, erosional outwash or the export of crop residues by harvesting (Dolan et al., 2006; Gregorich et al., 1998; Six et al., 2000). Therefore, grassland use correlates with higher inputs of OC whereas cropland use correlates with higher losses of OC. In Study I, a gradient from permanently used grasslands (highest stocks) to sites with alternations in grassland and cropping (medium stocks) to permanent cropland use (lowest stocks) was found. This illustrates the importance of the factor land use history in contrast to the actual land use at the time of sampling. The conversion from cropland to grassland may result in fast OC losses, whereas OC accumulation after conversion from grassland to cropland may be disproportionally slow ("slow in fast out") (Poeplau et al., 2011). In consequence, the current land use may not mirror the current OC status and alternating land uses may have an OC distribution associated with the dominant land use as seen in Study I. Authors in Belgium and the Netherlands showed that specific land uses such as plaggen manure, heathland farming or drainage may imprint OC stocks for large time periods (Meersmans et al., 2009; Schulp and Verburg, 2009; van Wesemael et al., 2010).

Wiesmeier et al. (2012) found land use to be a major controlling factor of OC storage analysing the whole soil profile (0 – 100 cm profile depth). The question arises to which soil depth the direct impact of this factor lasts. The importance of the predictor variable historical land use strongly decreased from 34% in the topsoil to 6% in the subsoil and no significant differences between subsoil OC stocks grouped by the historical land use were detected. Ward et al. (2016) found significant management effects on OC concentrations to 60 cm profile depth in English grassland soils. With respect to cropping systems, Hobley et al. (2018) detected OC gains in PK fertilised clover systems down to 1 m soil depth. However, the RF model of Study I implemented data on general land use and land use history, so data on specific management intensities or crop rotations within grassland or cropland use is absent in this study.

To summarise, Studies I, II and III have shown that the amount as well as the composition of topsoil OC stocks were determined by the legacy of land use, with highest OC stocks under constant grassland sites.

3.2 Controlling factors of subsoil OC storage

In Study I, the most important predictor variable of subsoil (30 – 100 cm profile depth) OC storage was soil type (40% relative variable importance) followed by major landform (27%) and TWI (12%). This resulted in highest subsoil OC stocks found in alluvial, colluvial and groundwater affected soils as well as in soils located in level and plain landform positions in Bavaria. In Study II, subsoil (30 – 120 cm profile depth) OC stocks gradually increased in a catena between landform positions from backslope to floodplain and again, floodplain subsoils contained significantly higher stocks than soils on slope. In both studies soils developed on sediments and soils affected by water saturation contained highest subsoil OC stocks. Both observations are closely related with the landform position and both studies disclosed Fluvisols as major subsoil OC reservoir.

Fluvisols as well as colluvial soils are soils that have developed on allochthonous sediments, i.e. fluvial deposits or material that has moved down a slope by gravitational action as a result of erosional wash (IUSS Working Group WRB, 2015). They deposit in floodplains (Fluvisols) or low-grade slopes and depressions (colluvial soils) and may contain high amounts of OC (Berhe et al., 2007; Doetterl et al., 2012a; Gregorich et al., 1998). Human activities such as clearing, tillage or the growing of erosion permitting crops such as Maize, can trigger the erosion of OC-rich topsoil and its lateral transport by water and wind (Leopold and Völkel, 2007; Montgomery, 2007; Pimentel and Kounang, 1998; VandenBygaart et al., 2012). The successive burial of these sediments can lead to the accumulation of large amounts of OC building up the subsoils of these soils (Chaopricha and Marín-Spiotta, 2014; D'Elia et al., 2017; Doetterl et al., 2012a). However, its OC content may be highly variable and depend on its source (Scott and Wohl, 2018). In the case of Study II, missing E layers of the Haplic Luvisols at backslope positions suggested that the soils have suffered from erosional mass losses. This process may have redistributed topsoil material and its associated OC to the lower lying footslope and toeslope of the catena. This hypothesis was supported by the low proportion of F^{14}C found in the subsoil OC in footslope positions, indicating that the photosynthetic fixation of this carbon was a substantial time ago. The stratification of toeslope and floodplain profiles was a strong indicator for the deposition of material, which may have originated upslope or upstream or a mixture of that. The relatively high proportion of $F^{14}C$ in the subsoils >30 cm depth of toeslope and floodplain OC indicated that the OC found in these sediments was photosynthetically fixed more recently. As a consequence, the sediments must have been deposited in more recent times than in footslope positions. Though, DOC transported by percolating slope water or groundwater may have shifted absolute ^{14}C contents and with that the $F^{14}C$ of these soils (Sanderman et al., 2009). This, in turn, suggested that old footslope subsoil OC was largely decoupled from the atmospheric carbon cycle.

Permanent or periodical water saturation, as the second emerging pedogenetic factor, typically affects the subsoils of Fluvisols and Gleysols. The resulting anaerobic conditions hamper the degradation of OM, which leads to the accumulation of OC in poorly drained soils (Berhe, 2012; Ponnamperuma, 1972; Reddy and DeLaune, 2008; Wiesmeier et al., 2012). Hydromorphic soil properties in low lying landform positions of Study II showed that soils in the floodplain as well as in the toeslope had been affected by groundwater or stagnating slope water. Water saturation and reduced oxygen availability most probably preserved once the deposited OC from decomposition (Blazejewski et al., 2005; Ponnamperuma, 1972; Reddy and DeLaune, 2008). Redoximorphic features, i.e. mottles, were found all over the floodplain and toeslope profiles of Study II. Strong reductive properties were only found in the deep subsoil at >100 cm profile depth. Together with the high absolute amounts of Fe_{ox} and increasing proportions of Fe_{ox} on Fe_{DCB} with depth, it was assumed that now mainly oxic conditions (± seasonal fluctuations) dominate in the uppermost 100 cm in these landform position and that the pedogenesis was relatively recent (Blume and Schwertmann, 1969; Lair et al., 2009a). A rapid accumulation of Fe oxides in Bt horizons after 30 years of drainage was also observed by Hayes and Vepraskas (2000). The present river morphology in combination with the findings on pedogenic oxides led to the assumption that artificial (weirs and ditches) and semi-natural (riverbed incision after regulations) drainage led to the oxidation of the uppermost 100 cm of the floodplain within few decades, which altered soil genesis. Such pedogenetic shifts from semi-terrestrial towards terrestrial systems in anthropogenically modulated floodplains are supported by Lair et al. (2009b), who reviewed the dynamics of European river-floodplain systems. Also, Veenstra and Lee Burras (2015) found human induced pedogenesis after 50 years of drainage of agricultural soils in Iowa. In contrast, low proportions of Fe_{ox} as well as intensive clay coatings in the subsoils of the Haplic Luvisols of footslope and backslope profiles indicated clay migration and therefore a relatively mature pedogenesis (Blume and Schwertmann, 1969). The soil type as a result of pedogenesis has also been identified as important OC factor in works by Suuster et al. (2012) and Wiesmeier et al. (2012). It can be argued if soil type is a truly independent OC predictor (Wiesmeier et al., 2019). As shown in Study II, the underlying processes that affected OC storage of the subsoils were associated with the reallocation and deposition of matter as well as alternations in soil hydrology. Both resulting in the genesis of very specific soil types that combine this variety of pedogenic properties. Hence, the classification of soil types is valuable for a deeper understanding of subsoil OC stocks.

Interestingly, no correlations were found between SOC stock and clay content, neither in Study I nor in Study II or III. The fine fraction particles (<20 µm) including 2:1 layered clay minerals and Fe oxides and hydroxides are known to stabilize OC (Hassink, 1997; Kleber et al., 2007; Mikutta and Kaiser, 2011). Also, clay content was found to be associated with the amount of OC stored as oPOM (Kölbl and Kögel-Knabner, 2004). Though, missing correlations between OC and clay in the presented studies may point to the fact that the soils were not saturated with OC. Wiesmeier et al. (2014a) showed the low degree of OC saturation in agricultural soils of Bavaria. This demonstrates that clay content is a weak predictor for OC storage and rather an indicator for the soils potential to store OC. Despite the low OC saturation, clay-rich soils resulted amongst the soil types with highest subsoil OC stocks in Study I.

To summarise, the findings demonstrated that topography and the generic soil type are closely linked together with respect to subsoil OC storage. In addition, the legacy of human activities not only amplified the lateral OC transport and therefore the amount of buried OC. Anthropogenically induced alterations of the river dynamics as well as drainage also accelerated pedogenesis within relatively few decades.

3.3 OC pools of floodplain subsoils

As shown in Study I and II, the subsoil OC was largely driven by the presence of OC-rich sediments as parent material and the degree of water saturation impeding composition. As a result floodplains were found to contain large OC reservoirs in their subsoils. Though, little is known about the quality of this OC.

The allocation of OC to particulate OM and organo-mineral associations was remarkably, as high proportions of this OC were found in light fractions. The contribution of mineral-bound OC to total OC decreased with depth and this was valid for the subsoils of both study sites of Study III. Relevant proportions of charred C were found in the $\mathrm{oPOM_{fine}}$ of the subsoils of both sites, as revealed by the molecular mixing model (Baldock et al., 2004). The presence of light fraction OM in general and of charred C in particular was explained by the floodplain's development on alluvial sediments and its variable composition with respect to the deposited POM (Noe and Hupp, 2009; Rennert et al., 2018). Agricultural burning practices are known to have increased the amount of pyrogenic C in European soils since the Neolithic (Gerlach et al., 2012; Knicker, 2011). The reduced vegetation after burning and the increase of arable land triggered soil erosion, which redistributed pyrogenic C from the hillslopes to the valleys (Gerlach et al., 2012) and finally its accumulation as alluvial deposits (Coppola et al., 2018; Cotrufo et al., 2016).

At the Otterbach site, subsoil $\mathrm{oPOM_{fine}}$ was largely depleted of easily decomposable carbohydrates and enriched with recalcitrant components such as lipids and proteins. In contrast, subsoil $\mathrm{oPOM_{fine}}$ at the Ammer site contained partially decomposed plant residues with high proportions of carbohydrates. Temporary fluctuations in the groundwater table in combination with high water-holding capacities due to silty-clay textured soils may have resulted in oxygen restrictions in the subsoil of the Otterbach site. This caused anaerobic microsites that favoured the preservation of aliphatic compounds (Keiluweit et al., 2016). In contrast, numerous worm casts and krotovinas in the subsoil of the Ammer site suggested high bioturbation, which provided fresh OM to the subsoil (Don et al., 2008; Lavelle, 1988). Therefore, it was assumed that the composition of POM was modulated by the specific site conditions in the subsoils after deposition.

In contrast to the light fraction OC, the organo-mineral associated OC (OMF_{fine}) was neither depleted nor enriched of any particular C-component, so it was assumed that its composition was independent of the specific site conditions or depth. No relationship between OMF_{fine} and fine texture (particles <20 µm) and no controls of clay mineralogy on OMF_{fine} were found. The OC coverage of OMF_{fine} was higher in all depths at the Ammer site compared with the Otterbach site. This observation was explained with abundant resources of Ca^{2+} and Mg^{2+} cations originating from the calcareous parent material at the Ammer site. As a result, cation bridges between clay minerals and the functional group of the OM led to higher OC coverages (Rowley et al., 2018).

To summarize, floodplain subsoils may contain relatively high amounts of light fraction OC. Depending on the specific site conditions (hydrologic regime, bioturbation) of the floodplain, this OC may be highly degraded or easily decomposable. The occurrence of charred C in floodplain soils was associated with the legacy of slash and burn practices, followed by erosion and deposition in alluvial sediments. The composition of the organo-mineral associated OC was not affected by any site condition. The presence of polyvalent cations led to higher OC coverages on mineral surfaces.

3.4 Key areas of OC storage in agricultural soils of Bavaria: Value and fate of floodplain soils

The three studies revealed temperate grassland floodplains as highly relevant reservoirs for topsoil and subsoil OC storage. The relevance of floodplain soils to store large amounts of OC is recognised by a growing number of studies (Cierjacks et al., 2010; D'Elia et al., 2017; Graf-Rosenfellner et al., 2016; Hoffmann et al., 2009; McCarty and Ritchie, 2002; Ricker et al., 2013; Steger et al., 2019; Zehetner et al., 2009). To illustrate, Wiesmeier et al. (2014b) estimated that floodplain soils stored 14% (109 Mt) of OC of the total OC stocks of Bavaria, occupying 11% of the total area. The question remains how vulnerable these OC stocks are to degradation and C losses. Due to the high OC content, floodplain soils are highly productive sites (Tockner and Stanford, 2002). Forty-six percent of the active floodplains in Germany are intensively used as grasslands and roughly 50% of the inactive floodplains, i.e. floodplains without regular inundations and located behind levees, are used as cropland (Brunotte et al., 2009). In a report about OC stocks of floodplains in Germany, Scholz et al. (2012) calculated that active floodplain soils under grassland use stored more than twice as much C than under cropland, i.e. 23 Mio. t C and 10 Mio. t. C respectively, which was about the magnitude observed in Study II. It is assumed that the conversion from grassland to cropland triggers the mineralisation of SOC (Guo and Gifford, 2002). So the question arises, if these large amounts of OC stored in floodplains are threatened by land-use changes. Krause et al. (2011) observed that floodplain meadows in Northern Germany were to large proportions transformed to intensively used grasslands since the 1950'ies. Xu et al. (2017) observed a general decrease of cropland and grassland on the one hand and on the other hand an increase of the total area of riparian forest in active floodplains of the upper Danube between 1963 and 2010. Concomitantly, the patch size of agricultural land increased, indicating the intensification of the land use. They concluded, that the less productive agricultural sites closest to the river were abandoned and changed to riparian forests, whereas highly productive sites at larger distance to the river were intensified including turning from grassland into cropland. With respect to the above discussed findings by Poeplau et al. (2011), this would have caused large OC losses in the short term. The German Federal Environment Agency (UBA) reported a general decline in permanent grassland sites of roughly 3% in Germany since 1991 (Bundesministerium für Umwelt, 2018). This trend slightly levelled off after 2013, when the EU's Common Agriculture Policy (CAP) started to financially support farmers for the maintenance of permanent grasslands and stop land-use changes ("greening measures") (Bundesministerium für Umwelt, 2018). However, these grants are optional, and economic incentives to cultivate corn and energy crops may increase the pressure to intensively cultivate the areas, including suitable floodplains.

Many floodplains have been intensively drained in the past in order to increase productivity and improve cultivation, but current data on the extent of drained floodplains in Germany and particularly in Bavaria are missing. As discussed above and seen in Study II, soils may respond within few decades to such modulations in their redox conditions. Meersmans et al. (2009) and van Wesemael et al. (2010) observed significant decreases in total OC storage after drainage of agricultural soils in Belgium. As seen in Study III, floodplain subsoils may contain large proportions of easily decomposable light fraction OC – partly occluded in aggregates as oPOM and partly as fPOM. In the case of drainage, these soils generally become more susceptible for decomposition of OM by heterotrophic microorganisms (Moyano et al., 2013). In the short term, fPOM is readily available for microbes as oPOM in aggregates is spatially not accessible (von Lützow et al., 2006). As shown by Cressey et al. (2018), floodplain subsoils are not limited to metabolically active populations but are capable of rapid mineralisation. However, the organic and inorganic agents that bind aggregates may not be permanent (Tisdall and Oades, 1982). Soil aggregates have a *life cycle* (Six et al., 2000) and their turnover releases formerly protected oPOM for decomposition (De Gryze et al., 2006). Alternations in the drying and wetting cycles induced by drainage measures may further destabilise the aggregates. Therefore, it can be assumed that in the short term particularly fPOM, but in the medium term also oPOM OC becomes vulnerable to decomposition.

An increasing number of floodplains in Europe is being restored (Szałkiewicz et al., 2018). This is a good approach from an ecologic perspective, as valuable natural habitats are being re-established (Samaritani et al., 2011). Nevertheless, river restoration also means that levees are being deconstructed and floodplain soils are destabilised. With that, floodplains are again incorporated into the river's dynamic evolution and bank erosion occurs (Florsheim et al., 2008; Ward and Stanford, 1995). Subsequent losses in soil OC might be then at least partly replaced by the deposition of woody debris (Cierjacks et al., 2010). The redistribution of soil inherent OC should nonetheless accounted for.

Christensen and Christensen (2003) as well as Kundzewicz et al. (2013) reported an increase in the magnitude and number of severe floods in Europe with climate change. This would also increase stream flow and sediment load of the rivers and finally the amount of exported OC from the floodplain (Glendell and Brazier, 2014; Oeurng et al., 2011; Wheater and Evans, 2009).

To summarise, temperate grassland floodplains are large OC reservoirs, which are potentially threatened to degradation. This is due to i.) economic pressures towards a more intensive use of the sites and concomitant land-use changes ii.) drainage measures that increase the oxidation of the floodplain's subsoils and with that the decomposition of light-fraction OM and iii.) erosional mass losses due to more extreme flooding events in a changing climate.

4. Conclusions

The three underlying studies of this dissertation demonstrated how closely tied the legacy of land use, landform position and the generic soil type are with respect to soil OC storage. The in-depth studies confirmed thereby the findings for total Bavaria. With respect to topsoil OC stocks, it was demonstrated that not only the amount of OC stored but also the chemical composition of topsoil OC was determined by the land use history. Highest amounts of OC stocks were found under constantly used grassland sites. The subsoil OC was determined by the interactions between landform position and generic soil type. The landform position, i.e. topographical factors, indicate the lateral transport of OC-rich topsoil due to erosion and its subsequent deposition in low lying positions. There, periodical or permanent water saturation and the concomitant reduced oxygen supply limits the microbial decomposition of OC. As a result of these processes, alluvial and colluvial soils contained highest subsoil OC stocks (Study I and II). The legacy of human activities such as slash and burn practices and tillage amplified the lateral OC transport and therefore the amount of buried OC in the alluvial and colluvial subsoils. This process was also mirrored in the detection of substantial amounts of aromatic C in the light fraction OM of the alluvial subsoils (Study III). It was furthermore demonstrated that human induced alterations of the catchment's hydrologic regime, such as river regulations and drainage, can accelerate the floodplain's pedogenesis within few decades by increasing bank erosion and the oxidation of the subsoil. This was indicated by the occurrence of redoximorphic features in the floodplain's soil profiles as well as the distribution of pedogenic Fe oxides of Study II.

Remarkably high amounts of the subsoil OC in the floodplain were stored in light fraction OM, i.e. free and occluded particulate organic matter. The composition of this light fraction OC depended on the composition of the respective alluvial sediments as well as on the hydrologic conditions in the floodplain. The specific site conditions led to large contrasts in the chemical composition of the oPOM: strongly decomposed, lipid-rich in less-aerated subsoils and weakly decomposed in well-aerated soils with high bioturbation. In well-drained floodplain soils, particularly the free POM was assumed to be vulnerable to decomposition in the short term, as no further stabilisation mechanism protects it from heterotrophic reduction. In the medium term, also the occluded POM may decompose as the turnover of aggregates releases OM. Floodplain OC stocks may not only be threatened by drainage. As floodplain soils are highly productive agricultural sites, the economic pressure of intensive cultivation and land-use changes from grassland to cropland are immanent. Also, erosional mass losses in the case of severe floods in a changing climate may cause losses of soil OC.

Soil type, landform and the legacy of land use were identified as most relevant, when disentangling the factors of topsoil and subsoil OC storage in agricultural soils of Bavaria. Floodplain soils were disclosed as major OC reservoirs, because of the dominant grassland use of these soils on the one hand (high topsoil OC stocks) and its development on OC-rich sediments on the other hand (high subsoil OC stocks). The presented dissertation demonstrated that these soils may be prone to the release of CO_2 as land-use changes and drainage may alter the decomposition of the floodplain's inherent OM. Human activities in the ecosystem of decades and centuries had a strong influence on the amount and composition of the OC that is today stored within the whole soil profile. This becomes particularly clear in Europe and Bavaria with its old cultural landscapes and soils.

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Appendix

Peer-reviewed publications (first-authored)

Mayer, S., A. Kühnel, J. Burmeister, I. Kögel-Knabner and M. Wiesmeier (2019). Controlling factors of organic carbon stocks in agricultural topsoils and subsoils of Bavaria. Soil & Tillage Research, 2019.192: p 22-32.

Mayer, S., A. Kölbl, J. Völkel, and I. Kögel-Knabner (2019). Organic matter in temperate cultivated floodplain soils: Light fractions highly contribute to subsoil organic carbon. Geoderma, 2019. 337: p. 679-690.

Mayer, S., D. Schwindt, M. Steffens, J. Völkel, and I. Kögel-Knabner (2018). Drivers of organic carbon allocation in a temperate slope-floodplain catena under agricultural use. Geoderma, 2018. 327: p. 63-72.

Oral presentations

Mayer, S., Kölbl, A., Völkel, J. and I. Kögel-Knabner (2019): Subsoil organic carbon dynamics in temperate grassland floodplains. European Geosciences Union General Assembly 2019, 07.-12.04.2019. Vienna / Austria.

Kriegs, S., Steffens, M., Schwindt, D. , Völkel, J., Kögel-Knabner, I. (2017): Zwischen Hang und Aue – Kohlenstoffdynamik im Einzugsgebiet des Otterbach. Jahrestagung der Deutschen Bodenkundlichen Gesellschaft 04.-06.09.2017. Göttingen.

Kriegs, S., Buddenbaum, H., Rogge, D., Steffens, M. (2015): imVisIR: Hochaufgelöste Klassifikation und Elementkartierung der Pseudovergleyung in Parabraunerdeprofilen. Jahrestagung der Deutschen Bodenkundlichen Gesellschaft 05.-10.09.2015. München.

PICO presentations

Kriegs, S., Hobley, E., Schwindt, D., Völkel, J., Kögel-Knabner, I. (2017): Exploring Soil Organic Carbon Deposits in a Bavarian Catchment. European Geosciences Union General Assembly 2017, 24.-28.04.2017. Vienna / Austria.

Kriegs, S., Buddenbaum, H., Rogge, D., Steffens, M. (2015): Digital soil classification and elemental mapping using imaging Vis-NIR spectroscopy: How to explicitly quantify stagnic properties of a Luvisol under Norway spruce. European Geosciences Union General Assembly 2015, 13.-17.04.2015 Vienna / Austria.

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Mayer, S., Kühnel, A., Burmeister, B., Kögel-Knabner, I., Wiesmeier, M. (2019): Controlling factors of organic carbon stocks in agricultural topsoils and subsoils: A case study from Bavaria, Southeast Germany. Food Security and Climate Change: 4‰ new tangible initiative for the soil. 17.-20.06.2019. Poitiers / France.

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Kriegs, S., Steffens, M., Schwindt, D., Völkel, J., Kögel-Knabner, I. (2016): Soil Organic Carbon Stocks in Depositional Landscapes of Bavaria. 2nd HEZagrar PhD Symposium. 26.04.2016. Freising.

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Kriegs, S., Buddenbaum, H., Rogge, D., Steffens, M. (2015): Digital soil classification and elemental mapping using imaging Vis-NIR spectroscopy: Quantification of stagnic properties in Luvisol profiles under Norway spruce and European Beech. 9th EARSeL SIG Imaging Spectroscopy workshop. 14.-16.04. 2015. Luxembourg.

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Other publications (co-authored)

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