

Beyond clay: towards an improved set of variables for predicting soil organic matter content

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Received: 19 December 2017 / Accepted: 27 January 2018 / Published online: 3 February 2018
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Abstract Improved quantification of the factors controlling soil organic matter (SOM) stabilization at continental to global scales is needed to inform projections of the largest actively cycling terrestrial carbon pool on Earth, and its response to environmental change. Biogeochemical models rely almost exclusively on clay content to modify rates of SOM turnover and fluxes of climate-active CO₂ to the atmosphere. Emerging conceptual understanding,

however, suggests other soil physicochemical properties may predict SOM stabilization better than clay content. We addressed this discrepancy by synthesizing data from over 5,500 soil profiles spanning continental scale environmental gradients. Here, we demonstrate that other physicochemical parameters are much stronger predictors of SOM content, with clay content having relatively little explanatory power. We show that exchangeable calcium strongly predicted SOM content in water-limited, alkaline soils, whereas with increasing moisture availability and acidity, iron- and aluminum-oxyhydroxides emerged as better predictors, demonstrating that the relative importance of SOM stabilization mechanisms scales with climate and acidity. These results highlight the urgent need to modify biogeochemical models to

Responsible Editor: Stuart Grandy.

Electronic supplementary material The online version of this article (<https://doi.org/10.1007/s10533-018-0424-3>) contains supplementary material, which is available to authorized users.

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better reflect the role of soil physicochemical properties in SOM cycling.

Keywords Soil organic matter · Biogeochemistry · Carbon cycle

Introduction

A clear description of soil organic matter (SOM) stabilization mechanisms is needed to inform society's ability to manage carbon and meet desirable climate targets (Minasny et al. 2017). An established and tested hypothesis is that clay content, i.e., the mineral fraction < 2 µm in size, principally determines SOM storage and stabilization by promoting the sorption of organic matter to mineral surfaces and aggregate formation (Oades 1988; Schimel et al. 1994; Wagner et al. 2007). Clay content may also alter SOM stability indirectly, by influencing microbial community composition, soil hydrology, drainage, and O₂ availability (Andrews et al. 2011; Fierer and Schimel 2002). The small size of clay particles imbues them with high specific surface area, and clay particles typically exhibit a combination of permanent structural charge and variable pH-dependent surface charge. The combination of charge and high surface area allows clay sized particles to dominate mineral–mineral, mineral–organic, mineral–metal, and mineral–water interactions in soils (Sposito et al. 1999). Thus, total clay content has been used as a proxy for the direct effects of aggregation and sorption, and indirect controls of soil water content on SOM stability (Kahle et al. 2003; Mikutta et al. 2006; Oades 1988). These insights are broadly applied in biogeochemical models (e.g., Coleman and Jenkinson 1996; Parton et al. 1987;

Wieder et al. 2015) in which clay or clay + silt content describes variation in SOM storage by modifying carbon fluxes into particular pools, and their rate of turnover. All of these models predict slower SOM turnover and higher SOM storage in more finely textured soils (Fig. S1). Although numerically tractable, these relatively simplistic considerations of SOM stabilization may not apply across diverse soil systems or capture the efficacy of different clay-sized particles for stabilizing SOM.

Current understanding suggests other physicochemical parameters may be better predictors of SOM content and stabilization. Observational and manipulative studies indicate that SOM content may be more related to the amount of extractable metals, here defined to include measures of exchangeable cations, pedogenic oxyhydroxides, short-range-order (SRO) phases, and organically complexed metals (Kaiser and Guggenberger 2000; Lawrence et al. 2014; Percival et al. 2000; Rasmussen et al. 2006; Torn et al. 1997). These studies highlight the fact that not all clay-sized particles are equivalent in their SOM stabilization capacity. The clay fraction includes some combination of phyllosilicates, crystalline and SRO oxyhydroxides and aluminosilicates, and organo-metal complexes, each of which vary in their surface area, reactivity, solubility, and sorptive capacity (Kaiser et al. 1997; Sposito et al. 1999). Furthermore, silicate clays are often coated with Al- and Fe-oxyhydroxides that contribute to their SOM stabilization capacity. Clay content as a sole measure can thus mask the diversity of mineral properties and may not effectively capture specific SOM stabilization mechanisms. It follows that other soil properties may serve as better surrogates of SOM stabilization than clay content alone.

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Recently, several attempts have been made to quantitatively link soil physicochemical variables with SOM abundance and stability across larger, regional spatial scales (Augustin and Cihacek 2016; Doetterl et al. 2015; Garrido and Matus 2012; Mathieu et al. 2015). In many of these studies, clay content was not a significant predictor of SOM content or stability, and climatic variables were identified as secondary in importance compared to soil mineralogical and morphological characteristics. Here, we address whether other soil physicochemical parameters are better predictors of SOM using a comprehensive set of soil profile data from the US National Cooperative Soil Survey Database that spans continental scale gradients in climates, ecoregions, and soil taxa.

Materials and methods

Global soil, climate, and ecoregion datasets

Soil data were compiled from the U.S. Department of Agriculture's National Cooperative Soil Survey (NCSS) Soil Characterization Database (<http://ncsslabdatamart.sc.egov.usda.gov/>). The data consist of 392,710 individual soil horizons sampled from 62,843 pedons described as part of NCSS activities. Data were assessed for quality assurance and control purposes by analyzing the distributions of each data type and errors corrected where possible or excluded when no obvious correction could be applied. Histosols and all organic horizons were excluded; only 240 horizons occurred in Gelisols and were not included in soil order specific analyses. The soil properties analyzed included SOC (%) used as a measure of SOM, horizon midpoint depth (depth; cm), clay (%), clay + silt (%), pH in H₂O, and extractable metals that included exchangeable Ca as determined by ammonium acetate extraction at pH 7.0 (C_{aex} ; $\text{cmol} + \text{kg}^{-1}$), dithionite extractable iron (Fe_d ; %), oxalate extractable iron (Fe_o ; %) and aluminum (Al_o ; %). The selected physicochemical variables were chosen to encompass a range of soil properties related to various proposed SOM stabilization mechanisms. This selection of variables also maximized the number of soil horizons that could be included in the analysis while maintaining a broad spectrum of soil properties. Soil organic carbon data were compiled following Wills et al. (2007), correcting total reported C for carbonate

C, and converting Walkley–Black organic carbon values to the equivalent of combustion C.

The soil data were paired with properties extracted from global maps of ecoregion, climate, and soil order. Ecoregion data were compiled from the World Wildlife Foundation terrestrial ecoregions map (Olson et al. 2001). Climate data, including mean annual temperature (MAT; °C), mean annual precipitation (MAP; mm year^{-1}), and potential evapotranspiration (PET; mm year^{-1}) were extracted from the 0.5 degree CRU climate dataset of New et al. (New et al. 1999). The climate data were used to calculate an aridity index for each location, defined as the ratio of annual PET/MAP, where ratios > 1 indicate water-limited systems and values < 1 indicate energy-limited systems. The aridity index data were further divided into seven humidity classes using hierarchical clustering, with the number of classes chosen to maximize the number of locations in each class and to correspond with climate zones. Soil order information was extracted from the NCSS database when available. For locations lacking taxonomic information, soil order was extracted from the NRCS Global Soil Region map available at: https://www.nrcs.usda.gov/wps/portal/nrcs/detail/soils/use/?cid=nrcs142p2_054013. Spatial data were handled using ArcMAP 10.3.1 (ESRI, Redlands, CA) and all statistical analyses performed using JMP Pro v12.1.0 (SAS Institute, Cary, NC).

Statistical analyses and model development

The data were filtered such that only horizons with a complete set of soil and climate variables were included, providing a total of 5587 pedons with 28,819 horizons. To standardize variation among variables and meet normality assumptions of the applied statistical tests, all continuous predictors and the soil organic carbon (SOC) data were transformed to normal distributions using Box-Cox transformations, followed by standardization to a mean of 0 and standard deviation of 1. The transformed and standardized data allowed for direct comparison of regression coefficient estimates among predictor variables.

Regression analyses

Linear mixed model (LMM) regression was implemented to determine the soil and climate parameters that best explained the variance in SOC content. LMM

was selected to address the non-independent nature of multiple horizons within one pedon. The models were structured with pedon ID as a random effect, allowing the slope and intercept of the relationship between soil depth and SOC to vary by each pedon based on the a priori knowledge of a relationship between soil depth and SOC content (Jobbagy and Jackson 2001). Three LMM models were run: (1) using just soil depth as a fixed effect, (2) using soil depth and the entire set of the selected continuous physicochemical and climate variables, including two-way interaction terms among all variables, as fixed effects, and (3) a reduced final model that included only those fixed effects that contributed the most to the prediction of SOC based on regression coefficient estimates and F-values. Specifically, the final model only kept terms with regression coefficient estimates > 0.1 or < -0.1 that corresponded with F-values > 400 . The fixed-effect variables in the final model included depth, Al_o , Ca_{ex} , pH, PET/MAP, and the pH by Al_o interaction term (Table 1). The relative importance and contribution of fixed effects was determined based on regression coefficient estimates and F values.

Additionally, LMM modeling of SOC was performed across pH classes. Soil-pH data were grouped using hierarchical clustering, with the number of classes chosen to maximize the number of samples in each class. We then ran LMM for each pH class, using pedon ID as the random variable and allowing both the slope and intercept to vary with depth, and included depth, Al_o , and Ca_{ex} as fixed parameters. The F-values for each fixed parameter were reported in Fig. 2b to demonstrate the relative importance of each parameter to account for SOC variance across pH classes. The relative contribution of each fixed parameter by pH class, in combination with soil physicochemical

property change by pH class (Fig. 2a), was used to develop the conceptual figure of change in dominant SOM stabilization mechanism with pH (Fig. 3).

Correlations by environmental categorical variables

The conditional residuals of the soil depth-only LMM were related to all soil physicochemical and climate-continuous variables, parsed among three categorical variables, using Spearman's ρ (Fig. 1). The categories were humidity class ecoregion, and soil order. This approach allowed us to include collinear variables left out of the LMM. Significance was evaluated based on a Bonferroni's corrected α value of 0.0002 ($\alpha = 0.05/250$ individual correlations). Only categories with $n > 400$ soil samples were used for correlation determination.

Results and discussion

Soil organic carbon and physicochemical parameters

Linear mixed model (LMM) regression indicated that soil depth was the strongest predictor of SOC, with a significant trend of decreasing SOC content with increasing depth. After accounting for depth, Al_o and Ca_{ex} were the best predictors, followed by pH, the interaction of pH and Al_o , and PET/MAP (potential evapotranspiration over mean annual precipitation). Al_o and Ca_{ex} showed a positive relationship with SOC, whereas pH, the pH- Al_o interaction term, and PET/MAP exhibited weaker, negative relationships with SOC (Table 1).

Table 1 Fixed-effect parameters from the linear mixed model

Term	Degrees of freedom	Regression coefficient	95% lower	95% upper	F ratio	Prob > F
Depth	5782	- 0.579	- 0.589	- 0.569	12,616	< 0.0001
Al_o	26,054	0.306	0.298	0.314	5135	< 0.0001
Ca_{ex}	25,934	0.342	0.330	0.354	3202	< 0.0001
pH	23,444	- 0.258	- 0.270	- 0.245	1725	< 0.0001
pH $\times$ Al_o	27,852	- 0.141	- 0.148	- 0.134	1492	< 0.0001
PET/MAP	6468	- 0.108	- 0.121	- 0.096	279	< 0.0001
Intercept	5198	- 0.022	- 0.036	- 0.009	–	–

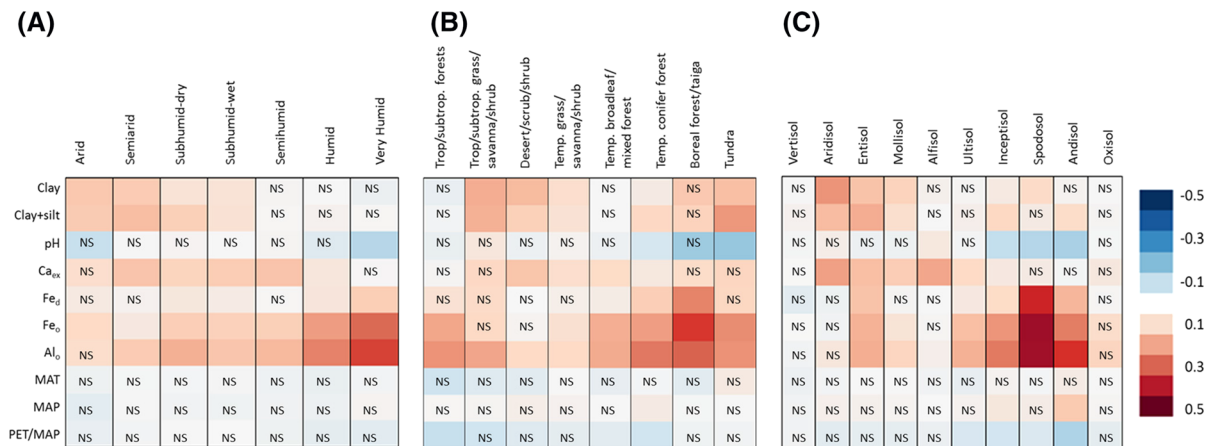


Fig. 1 Heat maps indicating the Spearman correlation between the conditional residuals of a mixed regression model using pedon ID as a random factor and horizon depth as a fixed effect, and soil physicochemical properties and climate parameters by the categorical variables of **a** humidity class, **b** ecoregion, and

c soil order. Spearman correlations are scaled from dark blue for negative correlations to dark red for positive correlations. Those correlations with $P > 0.0002$ are considered not significant (NS), based on Bonferroni's corrected α value

Correlation analysis of the conditional residuals from a LMM using only depth as the fixed effect was performed to further explore SOC relationships among the categorical variables of humidity class, ecoregion, and soil order (Fig. 1). There were few correlations of SOC with climate variables within each category; rather, a combination of either Fe_o and Al_o , or Ca_{ex} and clay or clay + silt best explained SOC. Exchangeable Ca and soil texture exhibited the strongest correlation in water-limited environments, whereas Al- and Fe-oxyhydroxides exhibited the strongest correlations in humid environments.

Among soil taxa, Entisols and Inceptisols, with minimal pedogenic development, presented strong positive correlations with nearly all soil properties (Fig. 1), likely owing to their large range of soil properties, and the diversity of environmental conditions in which these soils are found (Buol et al. 2011). At the other soil development and weathering extreme, no significant correlation to any soil property was observed for Oxisols. This may be due in part to the rapid litter and SOC decomposition associated with the warm, wet tropical climates typical of Oxisols (Tiessen et al. 1992). Oxisols are highly weathered and dominated by quartz and crystalline, variable charge Fe- and Al-oxides and kaolinite, with few to no weatherable minerals (Buol and Eswaran 2000). In these soils, Fe-oxide cementations form very stable concretions (El Swaify 1980) that serve as the

dominant mechanism for protecting SOM from decomposition (Tiessen et al. 1994). However, these concretions also confound soil textural analysis by resisting dispersion, biasing results towards a coarse classification while physically behaving more similarly to fine-textured soils. Taken together, these observations suggest a general decrease in the range of soil properties that can facilitate SOM stabilization with increased soil weathering (Lawrence et al. 2015; Torn et al. 1997).

Prevalence of extractable Ca and texture in dry climates

The regression and correlation analyses reflected strong positive correlation of SOC to Ca_{ex} , clay + silt, and clay in water-limited systems, that included deserts, grasslands, savannas, and shrublands, largely characterized by Aridisols, Mollisols and Alfisols (Fig. 1). These ecoregions with less precipitation maintain circum-neutral to alkaline pH values, and retain non-hydrolyzing base cations. These conditions favor the formation of 2:1 phyllosilicates with an abundance of negatively charged exchange sites (Douglas 1989; Kittrick 1971). When divalent ions, such as Ca^{2+} , occupy these exchange sites, they can bind negatively charged organic moieties, such as the carboxylic acid functional group common on SOM, effectively forming a bridge with the underlying clay

(Mikutta et al. 2007). Such cation bridging is widely recognized as an important SOM stabilization mechanism (von Lützow et al. 2006). In addition, exchangeable Ca can cross-link two negatively charged organic functional groups together, causing SOM aggregation and immobilization (Kunhi Mouvenchery et al. 2012). Calcium has a stronger influence on SOM stabilization in dry climates than other divalent cations because of its higher relative abundance (Smith et al. 2014) and greater ionic radius, which facilitates stronger binding energies (Muneer and Oades 1989). The strong correlation of SOC to exchangeable Ca and texture in water-limited systems is consistent with previous work highlighting a central role for texture in modulating SOM cycling (Amato and Ladd 1992; Burke et al. 1989; Hassink 1997; Nichols 1984), work that served as the foundation for current biogeochemical models.

Prevalence of extractable Al and Fe in humid climates

Soil carbon content in humid, forested ecoregions (generally represented by Inceptisols, Ultisols, Andisols, and Spodosols) exhibited strong positive correlations with Fe_o and Al_o, a negative correlation with pH, and little to no correlation with clay or clay + silt (Fig. 1). In these systems, the combination of water availability, an abundance of low molecular weight organic acids, or distinct primary mineral assemblages, facilitates high rates of mineral weathering that may release Fe, Al, and Si faster than crystalline minerals can precipitate (Shoji et al. 1993). This can lead to soil solutions that are oversaturated with respect to SRO phases, that once formed, have high surface areas and high densities of reactive hydroxyl sites (Harsh et al. 2002), allowing them to immobilize large quantities of SOM. These SRO phases have also been implicated as important drivers of aggregation, which further protects SOM against decomposition (Asano and Wagai 2014; Rasmussen et al. 2005). The generally high SOM content in humid systems also promotes Al and Fe-organic complexation and co-precipitation of organo-metal complexes, which can compete and occur in conjunction with SRO formation (Dahlgren et al. 2004). Both phases are important to SOM stabilization (Wagai and Mayer 2007), but the analyses herein were based on SOC correlations with oxalate-extractable Al or Fe, which includes both

organo-metal complexes and SRO phases, so we are not able to directly separate these factors. We did not include more direct measures of organo-metal complexation because of the limited extent where it was measured. However, as indicated below (Fig. 2), for those locations with such measures, we observed strong gradients in the relative concentration of organo-metal complexes and SRO phases with soil pH.

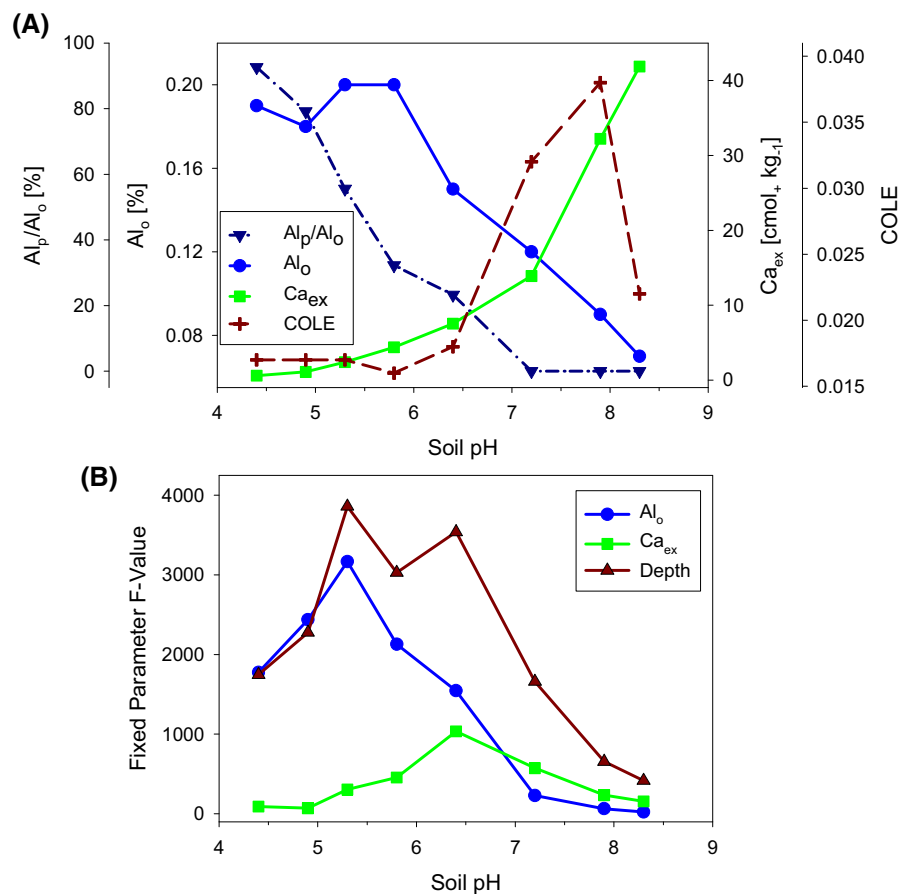
Soil pH and stabilization mechanisms

Our results suggest a complicated and important influence of soil pH on SOM content. The LMM results indicated a significant role for pH, and the interaction of pH and Al_o in accounting for SOC content, but pH was not significantly correlated with SOC across most categories. Soil pH reflects the overall chemical state of the soil system and dictates a number of geochemical gradients, including the speciation of dissolved metals, the reactivity and charge of minerals and organic molecules, and the predominant type of organo-mineral bond (Deng and Dixon 2002). Each of these influences SOM stabilization, resulting in an overall influence of pH that is nonlinear and driven by thresholds in aqueous speciation and mineral stability, which are in turn modulated by water availability and other soil-forming factors.

Soil pH corresponded with distinct gradients in soil properties (Fig. 2a). In particular, Ca_{ex} increased significantly above pH 6.5, whereas Al_o was greatest at pH values < 6.5. Although there were not enough data to include them in the full analysis, two additional values, the coefficient of linear extensibility (COLE), and pyrophosphate extractable Al (Al_p) were summarized by pH class to further elucidate soil property changes with pH. COLE provides a measure of soil shrink-swell capacity that corresponds with clay content and presence of expansible 2:1 phyllosilicates; Al_p is a common metric for organically complexed Al. COLE increased above pH 6.5, whereas the ratio Al_p/Al_o, which provides an estimate of the proportion of organo-metal versus organo-mineral Al species, increased at pH values less than 5.5, with Al_p dominant at low pH.

Regression analyses across pH classes (Fig. 2b), using Al_o to represent SRO phases and organo-metal complexes and Ca_{ex} to represent 2:1 clays and

Fig. 2 a The change in soil physicochemical properties important for predicting SOC content by soil-pH class, including the soil content of oxalate extractable Al (Al_o), the fraction of Al_o that is partitioned to Al- organo-metal complexes as measured by Na-pyrophosphate extractable Al (Al_p), the amount of Ca on the cation exchange complex (Ca_{ex}), and the coefficient of linear extensibility (COLE) (parameter values are medians within each pH class). **b** Fixed parameter F-values for Al_o , Ca_{ex} and horizon depth from linear mixed effect models using pedon ID as the random effect performed for each pH class



exchangeable cations, indicated that, after accounting for soil depth, Ca_{ex} was the most important factor at $pH > 6.5$, and Al_o was the dominant variable at $pH < 6.5$. The Ca-bridging mechanism is particularly important in soils enriched in 2:1 phyllosilicates (Laird 2001), corresponding with the increase in COLE at $pH > 6.5$ (Fig. 2a). The dominance of Al_o and the increase in the Al_p/Al_o noted at $pH < 5.5$ (Fig. 2a) suggest that SRO is the dominant factor at pH 5.5–6.5, whereas at low pH, organo-metal complexation is the dominant factor controlling SOC content (Wagai and Mayer 2007).

Here, we propose a conceptual model whereby the dominant SOM stabilization mechanism varies as a function of pH (Fig. 3). The SOM stabilization mechanisms vary with increasing soil pH from predominantly organo-metal complexation, to association with SRO phases, to Ca complexation and cation bridging with phyllosilicates. Soil pH varies nonlinearly with soil depth and soil-forming factors

(Fig. S5), and soil physicochemical properties vary nonlinearly with pH (Fig. 2a), leading to a complex and indirect relationship between pH and SOM content.

Implications for model improvement

Our analyses demonstrated that variables other than clay content serve as better predictors of SOM content in many systems. This observation implies that models for continental to global scale SOM content may be improved by inclusion of alternate mineralogical proxies. Variables of particular importance include exchangeable Ca, SRO Al- and Fe-oxyhydroxides, and Al-, Fe- organo-metal complexes, and the relative contribution of each parameter can be scaled with pH (Fig. 3). A step to improve models would be to develop explicit functions that relate these variables to specific rate and transfer coefficients.

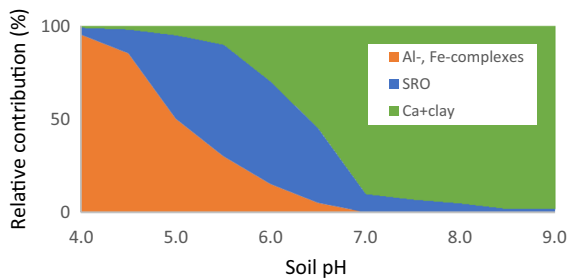


Fig. 3 Proposed conceptual model of how horizon-level soil pH controls the relative importance of SOM stabilization mechanisms grading from Al- and Fe- organo-metal complexes in acidic systems, to relative dominance by short-range-order (SRO) materials in moderately acid soils, to exchangeable calcium and clay content in circum-neutral to basic soils. The conceptual model is based on results of mixed regression modeling of soil organic carbon content across pH classes

For broad applicability in Earth system modeling, a new set of global-scale data products are needed that include the variables identified in our analysis. Currently, only data products on clay content and pH are available at the global level (Hengl et al. 2017), but as this analysis shows, extractable metal data do exist for a wide range of locations and soil taxa, and some globally gridded information about soil mineral composition has been derived (Ito and Wagai 2017; Journet et al. 2014). Given the large number of soil profiles that contain these data, compiling this information into large-scale data products should be a priority. With the current state of soil data products, global grids of soil pH may provide the most immediate and tractable variable for modifying biogeochemical models and their treatment of different SOM stabilization mechanisms (Fig. 3). Implementing such changes would strengthen the relationships among our conceptual understanding of SOM stabilization, model design and structure, and common measurements of soil physicochemical properties.

Acknowledgements This work was conducted as a part of the “What Lies Below? Improving quantification and prediction of soil carbon storage, stability, and susceptibility to disturbance” Working Group supported by the John Wesley Powell Center for Analysis and Synthesis, funded by the U.S. Geological Survey. Additional support was provided by NSF EAR-1331408 and EAR-1123454 to Rasmussen, NSF CAREER BCS-1349952 to Marin-Spiotta, US Department of Agriculture NIFA 2015-67003-23485 and US Department of Energy TES DE-SC0014374 to Wieder, and USDA-NIFA Hatch project HAW01130-H to Crow. Any use of trade, firm, or product names is for descriptive purposes only and does not imply endorsement by the U.S. Government.

Author Contributions This work is the result of two workshops sponsored by the USGS John Wesley Powell Center for Analysis and Synthesis in May of 2016 and May of 2017. The motivation and ideas for this work were generated collaboratively among all authors during these workshops. Rasmussen led manuscript development, data compilation, and analysis. All authors contributed to writing/editing, statistical analyses, and figure development.

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