



1 **A spatially explicit dataset of agriculture liming across the contiguous United**  
2 **States**

3 Samuel Shou-En Tsao<sup>1</sup>, Tim Jesper Surhoff<sup>2,3</sup>, Giuseppe Amatulli<sup>1</sup>, Maya Almarez<sup>1</sup>,  
4 Jon Gewirtzman<sup>1</sup>, Beck Woollen<sup>3</sup>, Eric W. Slessarev<sup>4</sup>, James E. Saiers<sup>1</sup>, Christopher  
5 T. Reinhard<sup>6</sup>, Shuang Zhang<sup>5</sup>, Noah J. Planavsky<sup>3</sup>, Peter A. Raymond<sup>1</sup>

6

7 <sup>1</sup> School of the Environment, Yale University, New Haven, CT 06511, USA

8 <sup>2</sup> Yale Center for Natural Carbon Capture, Yale University, New Haven, CT 06511, USA

9 <sup>3</sup> Department of Earth and Planetary Sciences, Yale University, New Haven, CT 06511, USA

10 <sup>4</sup> Department of Ecology and Evolutionary Biology, Yale University, New Haven, CT 06511, USA

11 <sup>5</sup> Department of Oceanography, Texas A&M University, College Station, TX 77843, USA

12 <sup>6</sup> School of Earth & Atmospheric Sciences, Georgia Institute of Technology, Atlanta, GA 30332, USA

13 *Correspondence to:* Samuel Shou-En Tsao (samuel.tsao@yale.edu)

14

15

16

17

18

19

20

21

22

23

24

25

26

27

28



## 29 Abstract

30 Agricultural lime has historically been applied to croplands in the United States to  
31 counteract soil acidification and enhance soil fertility, with important consequences  
32 for crop productivity and Earth's carbon cycle. Previous work on agricultural liming  
33 has largely focused on either region-specific case studies or national-level estimates  
34 of total application rates, leaving a major gap in understanding the spatial variability  
35 in lime application. This study addresses this gap by presenting the first spatially  
36 explicit dataset of agricultural lime application across the contiguous United States.  
37 The dataset comprises state-level data for 1930–1950 and a more detailed county-  
38 level dataset for 1954–1987, enabling comprehensive spatial-temporal analyses at  
39 multiple scales. Counties in the Midwest region exhibited the highest total amounts of  
40 lime applied in the latter half of the twentieth century, reflecting intensive agricultural  
41 activity. These counties were characterized by higher overall lime application rates  
42 (amount of lime applied per unit of limed area each year) but relatively lower liming  
43 frequency (ratio of limed area to total agricultural land area each year). In contrast,  
44 counties in the southeastern coastal region exhibited lower lime application rates per  
45 unit of limed area but more frequent lime applications. We used a machine learning  
46 framework, to elucidate key environmental and agricultural drivers of lime  
47 application. Our results show that the total amount of lime applied, as well as the  
48 application rate and frequency, are strongly associated with regional climatic  
49 conditions and soil properties. However, we also found evidence that agricultural  
50 management practices (such as crop production, fertilizer use, and soil pH  
51 recommendations) played a key role in shaping liming applications. Spatiotemporal  
52 integration of the data product results in a revised national estimate of total lime  
53 application, with a range of 15–25 million tons (Mt) per year. This study establishes a  
54 critical observational baseline for assessing the potential of agricultural lime  
55 application as a climate mitigation strategy and highlights the need for further  
56 research into its long-term environmental impacts.

57



## 58 1 Introduction

59 Agricultural lime has been historically applied to the surface of cropland to enhance  
 60 soil pH and increase yields (Tennant, 1799; Johnston, 1849). Soils can naturally  
 61 acidify through a range of processes, including atmospheric deposition of strong  
 62 acids, nitrification, sulfur oxidation, and release of organic acids by roots,  
 63 microorganisms, and decomposing organic matter (Goulding, 2016). In croplands,  
 64 soil acidification is intensified by repeated addition of nitrogen fertilizers, which  
 65 promotes the release of hydrogen ions ( $H^+$ ) via nitrification and by removal of base  
 66 cations in crop biomass (Guo et al., 2010). Approximately 50% of global arable land  
 67 is considered acidic ( $pH < 5.5$ ) (von Uexküll & Mutert, 1995). As soil pH decreases,  
 68 excess hydrogen ions replace base cations such as calcium ( $Ca^{2+}$ ) and magnesium  
 69 ( $Mg^{2+}$ ), causing their leaching and leading to a relative abundance of toxic elements  
 70 like aluminum ( $Al^{3+}$ ), which can be detrimental to crop growth (Fageria & Baligar,  
 71 2008). In response, where access is available lime application has become a common  
 72 strategy to mitigate the issue of soil acidification in croplands where access is  
 73 available. When lime is applied to soil, the  $Ca^{2+}$  and  $Mg^{2+}$  it releases can displace  $H^+$ ,  
 74 effectively reducing soil acidity (Hooda & Alloway, 1996). Repeated nitrogen  
 75 application and harvest increase soil acidity, prompting periodic lime applications to  
 76 restore soil pH towards neutral ( $pH \sim 7$ ). In addition to counteracting soil pH decline,  
 77 liming also replenishes key nutrients like  $Ca^{2+}$ ,  $Mg^{2+}$ , and potassium (K) (Han et al.,  
 78 2019), improves soil fertility and crop yield (Pagani & Mallarino, 2012), and has  
 79 become an essential practice in agriculture management in the U.S. (Agegnehu et al.,  
 80 2021).

81 When lime is applied to soils, it can be weathered by strong acids, such as nitric  
 82 acid ( $HNO_3$ ) produced from fertilizers, or by the weaker carbonic acid ( $H_2CO_3$ ),  
 83 which may originate from the dissolution of atmospheric  $CO_2$  or respired organic  
 84 carbon (Semhi et al., 2000). The relative contribution of strong acid derived acidity  
 85 and carbonic acid depends on the pH of the soil (Plummer & Wigley, 1976).  
 86 Depending on soil pH, the bicarbonate ion produced from weathering also, through  
 87 equilibration of the carbonic acid system, converts to carbonic acid and leads to  $CO_2$   
 88 source to the atmosphere. At higher pH values, however, carbonate weathering can  
 89 act as a  $CO_2$  sink, even on short timescales. Historically, lime application has been  
 90 assumed to be a net source of  $CO_2$  to the atmosphere, based on the premise that the  
 91 majority of lime is weathered by strong acids (Robertson et al., 2000; De Klein et al.,  
 92 2006). However, West & McBride (2005) estimated that only 38% of lime is  
 93 dissolved by strong acids. Subsequent studies have revised this estimate to be even  
 94 lower, with reported fractions ranging from 14% (Oh & Raymond, 2006) to 25%  
 95 (Hamilton et al., 2007). These findings suggest that most of the lime reacts with  
 96 carbonic acid ( $H_2CO_3$ ), forming bicarbonate ( $HCO_3^-$ ) in soil water. Nonetheless,  
 97 previous estimates are derived from conceptual models or localized studies,  
 98 underscoring the need for a spatially explicit dataset on agricultural lime applications  
 99 to better quantify the fraction of strong acid weathering and the carbon removal



100 potential of lime across diverse regions.

101

102 The application of crushed rocks to agriculture fields, referred to as enhanced rock  
 103 weathering (ERW), has emerged as a promising carbon dioxide removal and climate  
 104 mitigation strategy (Beerling et al., 2020). Current ERW trials predominantly  
 105 emphasize silicate rocks, in part because carbonate amendments like lime have  
 106 historically been viewed as net CO<sub>2</sub> sources. However, carbonates weather  
 107 significantly faster than silicates and contribute substantially to carbon cycling and  
 108 sequestration on shorter timescales (<3,000 years) (Gaillardet et al., 1999; Liu et al.,  
 109 2011). Enhanced carbonate weathering, particularly through large-scale agricultural  
 110 liming, could offer a promising and more immediate approach to atmospheric CO<sub>2</sub>  
 111 mitigation given its current widespread use. Using modelling approaches, Zeng et al.  
 112 (2022) estimated that application of lime to its maximum extent globally could  
 113 potentially sequester up to ~ 0.9 Gt of carbon per year, and Zhang et al. (2022) shows  
 114 that rivers have a generally high a capacity to transport this additional sequestered  
 115 carbon to the ocean (Knapp & Tipper, 2022). Furthermore, the application of lime has  
 116 been shown to reduce the emission of nitrous oxide (N<sub>2</sub>O) and methane (CH<sub>4</sub>)  
 117 emissions from soils (Khaliq et al., 2019; Shaaban et al., 2020), due to changes in the  
 118 biotic and abiotic environment that are tied to soil pH (Zhang et al., 2022). Since lime  
 119 can act as both a carbon source and a sink for atmospheric CO<sub>2</sub>, and its carbon  
 120 footprint can shift over time, often transitioning from a source to a sink depending on  
 121 environmental and management conditions, it will be important to develop spatial  
 122 understanding of where it can be marketed as a robust carbon removal practice. A  
 123 more detailed understanding of historical lime applications across regions would serve  
 124 as a valuable observational baseline to understand its climate impact in different  
 125 spatial regimes and further explore the potential of enhanced lime application as a  
 126 climate solution.

127 West & McBride (2005) provided the first national estimate of agricultural lime  
 128 applied in the continental U.S., reporting a peak of approximately 35 Tg C/year  
 129 during the 1970s. However, their data included only a time-series of national total  
 130 estimates, lacking spatial detail. To address this research gap, this study aims to

- 131 1. Compile a spatially explicit liming dataset (state-level for the period 1930–1950  
 132 and at the county-level for 1950–1990) for the U.S.
- 133 2. Use machine learning approaches to identify key agricultural and environmental  
 134 factors that explain the spatial variation in lime applications.

135 We find that liming has historically occurred almost exclusively in the eastern half of  
 136 the United States, which is characterized by more acidic soils. The spatial variation of  
 137 lime application has been further shaped by variation in agricultural practices,  
 138 climate, and soil properties. Our spatially explicit, multi-decadal dataset enables  
 139 improved assessment of liming's agricultural and climate impacts across the U.S.



## 141 2. Data and Methods

### 142 2.1 Liming data

143 We compiled state-level liming data from 1930 to 1950 (Mehring et al. 1957) and  
 144 county-level liming and agricultural data from 1954 to 1987 (U.S. Bureau of the  
 145 Census, 1946, 1956, 1961, 1967, 1977, 1981, 1984, 1989) as tabulated within ICPSR  
 146 dataset at the University of Michigan (Haines et al. 2018;  
 147 <https://www.icpsr.umich.edu/web/ICPSR/studies/35206>). Liming variables, including  
 148 ‘lime (tons)’ and ‘lime (acres)’, were only available for the years 1954, 1959, 1964,  
 149 1969, 1974, 1978, 1982, and 1987. In each census years, there are some counties that  
 150 did not report liming-related data. We will address the treatment of these missing  
 151 values in the following section on spatiotemporal interpolation. Lime application data  
 152 was not reported in Census records prior to 1954, and after 1987, it was aggregated as  
 153 ‘area of fertilizer and lime application’. Our dataset nevertheless covers a critical  
 154 period when national agricultural lime application reached its peak during the 1970s  
 155 (West & McBride, 2005). We convert all the units of liming variables to metric tons  
 156 (t) and hectares (ha) in the calculations, visualizations, and final data output.

### 157 2.2 Environmental and agronomic variables

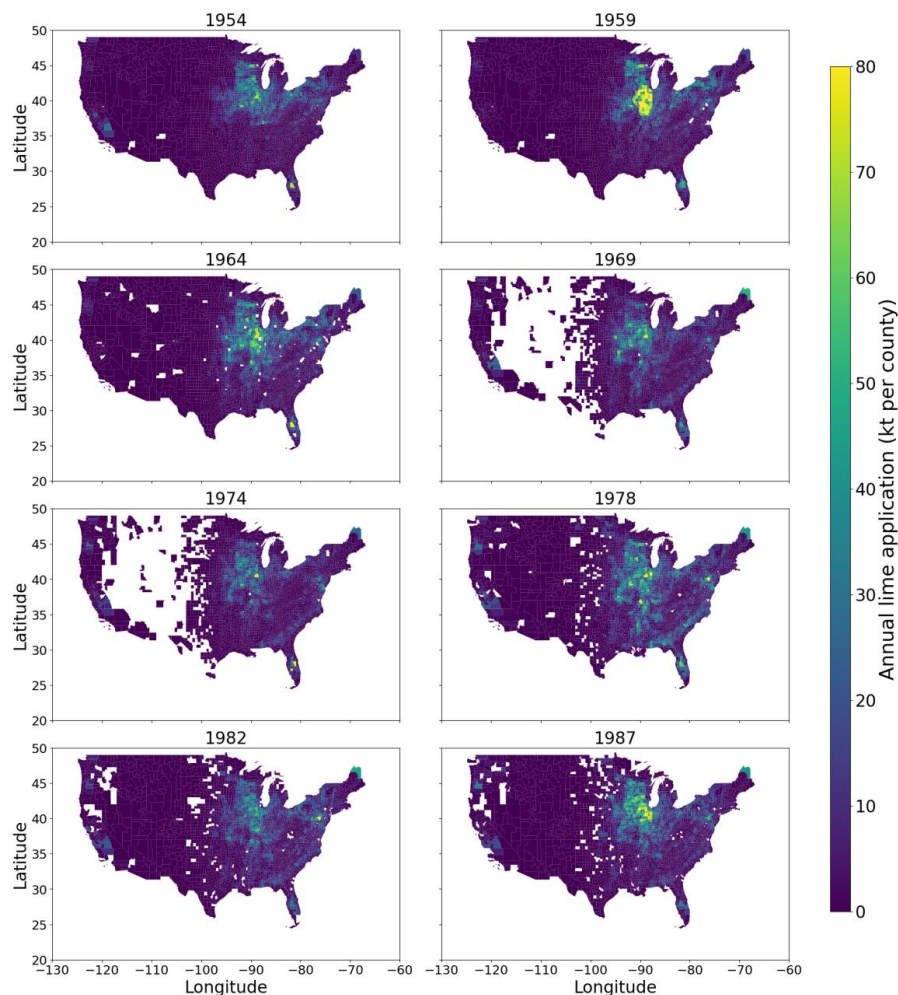
158 We compiled a comprehensive suite of environmental parameters, including  
 159 agricultural metrics, climate conditions, soil properties, and lithological features, to  
 160 further understand the potential controls of the spatial variation of agricultural liming  
 161 (Table S1). Agricultural data on specific crop production (e.g., corn, soybeans, wheat,  
 162 cotton) and land use (e.g., cropland harvested, cropland pastured, irrigated acreage)  
 163 were also collected from the Census reports. County-level nutrient data from  
 164 fertilizers and manure are extracted from Falcone (2021)  
 165 (<https://pubs.usgs.gov/publication/ofr20201153>). Averaged precipitation and  
 166 temperature data (1978-1987) were sourced from the PRISM Climate Group ([PRISM](https://prismclimate.org/)  
 167 [Climate Group, Oregon State University, 2014](https://prismclimate.org/); accessed March 20, 2025). Soil  
 168 properties (i.e., cation exchange capacity and soil carbonate stocks as  $\text{CaCO}_3$   
 169 equivalents) were obtained from re-gridded SSURGO data obtained from  
 170 Walkinshaw et al. (2023) (<https://casoilresource.lawr.ucdavis.edu/soil-properties/>).  
 171 Lithological data, specifically the occurrence of carbonate sedimentary rocks, were  
 172 sourced from Hartman & Moosdorf (2012) ([https://www.geo.uni-](https://www.geo.uni-hamburg.de/en/geologie/forschung/aquatische-geochemie/glim.html)  
 173 [hamburg.de/en/geologie/forschung/aquatische-geochemie/glim.html](https://www.geo.uni-hamburg.de/en/geologie/forschung/aquatische-geochemie/glim.html)), as these rocks  
 174 are assumed to weather more rapidly than silicates and thus influence soil acidity. We  
 175 adapted the county-level soil pH recommendation from a state-level agronomic  
 176 extension of pH recommendations based on the most commonly grown crop in each  
 177 U.S. county using CroplandCROS (<https://croplandcros.scinet.usda.gov>). Raster-  
 178 format datasets were spatially aggregated by averaging pixel values within the  
 179 geographic boundaries of each county. This process assigns a single representative  
 180 value to each county, ensuring that all raster-based variables are consistently aligned  
 181 with the county-level analysis. The agricultural data corresponds to the specific years



182 of the lime application records, while the climate, soil, lithology, and pH  
 183 recommendations lack temporal resolution and are treated as static across all years.  
 184 To help explain the temporal variability observed across different years, we also  
 185 extracted information on fertilizer application area and costs from the Census reports,  
 186 as well as farm expenditures and income.

### 187 **2.3 Spatial temporal interpolation**

188 For visualization and analysis, the county-level data were joined with the U.S.  
 189 county-level shapefile (<https://www.census.gov/cgi-bin/geo/shapefiles/index.php>).  
 190 We excluded counties outside the contiguous United States (e.g., Alaska, Hawaii). In  
 191 each census year, there were some counties that did not report data on lime  
 192 application. The counties with missing data for each year (Fig. 1) were addressed  
 193 using linear interpolation based on values from the preceding and following census  
 194 years within the 1954–1987 time range. In cases where data were available only for  
 195 the preceding year (e.g., the final year in the time series), missing values were  
 196 extrapolated using the previous value. After applying temporal interpolation, a  
 197 number of independent cities in the shapefile, including Baltimore, St. Louis, and  
 198 several independent cities in Virginia, remain absent from the census reports. These  
 199 cities are administratively distinct from counties in the U.S. Census framework and  
 200 are not included in county-level reporting. Consequently, no lime application data is  
 201 available for them across any census year. Given that these independent cities are  
 202 highly urbanized, with little to no agricultural land, it is reasonable to assume that  
 203 lime was not applied in these areas (see SI for further details on data processing).  
 204 Notably, most counties missing original lime application data are situated in areas  
 205 with limited agricultural activity, particularly in the arid and desert regions of the  
 206 western United States (Fig. 1). Since these missing gaps are not located in major  
 207 liming regions, the total lime application estimates are only slightly affected by the  
 208 number of missing counties each year and the interpolation procedure (Fig. S1).



209  
 210 Figure 1 Lime application (kilotons, kt) by county in the continental U.S. from 1954 to 1987.  
 211 The white gaps indicate counties that originally reported missing data, before temporal  
 212 interpolation and spatial filling were applied. The eight maps correspond to the census years  
 213 between 1954 and 1987.

214 We conducted the data processing mentioned above (temporal interpolation and  
 215 spatial gap filling) for both lime applied weight (kt) (Fig. 4) and limed area (ha) (Fig.  
 216 5) and all the other agricultural metrics (i.e., cropland area, crop production, nutrient  
 217 input) for each county. This resulted in a complete dataset with no missing value,  
 218 enabling us to conduct further derivation and statistical analysis. We were then able to  
 219 derive the lime application rate (lime applied weight/limed area) and liming frequency  
 220 (limed area/cropland area) and further analyze the main environmental controls on  
 221 each variable.

222





## 223 2.4 Statistical analysis

### 224 2.4.1 Pearson correlation and variable selection

225 To investigate the key factors controlling the spatial distribution of lime  
 226 application, we used the average across three census years (1977, 1982, and 1987),  
 227 representing the most recent decade with overlapping liming and environmental data.  
 228 This period offers a more stable and representative snapshot of liming conditions and  
 229 is closer to modern practices. We began with a comprehensive set of agronomic, soil,  
 230 and climatic variables (Table S1) and conducted a Pearson correlation analysis to  
 231 evaluate linear relationships between each predictor and lime application. To mitigate  
 232 multicollinearity, we also examined pairwise correlations among predictors and  
 233 removed variables from highly correlated pairs (Pearson's  $r > 0.90$ ), including  
 234 *Harvested crop land (acres)* vs. *Total Cropland (acres)* ( $r = 0.96$ ), *N fertilizer (kg)* vs.  
 235 *P fertilizer (kg)* ( $r = 0.91$ ), and *N manure (kg)* vs. *P manure (kg)* ( $r = 0.98$ ). Based on  
 236 domain relevance, data availability, and predictive strength, we retained one variable  
 237 from each pair and removed the others. Additionally, we calculated the Variance  
 238 Inflation Factor (VIF) for all remaining predictors. Variables with high VIF values  
 239 were selectively removed to reduce multicollinearity and improve model stability.  
 240 The final list of features also excluded: *Harvested crop land (acres)*, *P manure (kg)*, *P*  
 241 *fertilizer (kg)*, *Silt (%)*, *Total Cropland (acres)*, and *Available Water Storage (cm)*.  
 242 This process yielded a final set of 32 predictors with acceptable VIF values (range:  
 243 1.12 to 7.32), which were used for statistical and machine learning modeling (see SI,  
 244 Table S2). This step ensured a more robust and interpretable analysis of  
 245 environmental controls.

### 246 2.4.2 Random Forest Analysis

247 We employed a machine learning technique to account for complex relationships  
 248 between lime applications and the predictor variables. Random Forest (RF) is a  
 249 commonly used machine learning algorithm that can be used in both classification or  
 250 regression by constructing an ensemble of decision trees to capture complex  
 251 interactions and non-linear dependencies between predictor variables and the target  
 252 variable (Breiman, 2001). This approach is particularly suited for modelling in an  
 253 environmental or agricultural context characterized by high-dimensional,  
 254 heterogeneous relationships (Shen et al., 2020; Burdett & Wellen, 2022; Siqueira et  
 255 al., 2024; Jeong et al., 2016). In the context of our study, the RF model is not intended  
 256 to extrapolate temporal trends beyond the observed data range. Instead, it is used to  
 257 identify and quantify the relative importance of key environmental and agricultural  
 258 factors that may influence lime application patterns.

259 We implemented the model using the RandomForestRegressor class derived from  
 260 bootstrapped datasets (Pedregosa, 2011). We performed a grid search across multiple  
 261 combinations of hyperparameters using 5-fold cross-validation. Specifically, we  
 262 tested: number of trees ( $n\_estimators$ ) = 100, 300, 500; maximum depth ( $max\_depth$ )  
 263 = 10, 20, and unlimited (None); minimum samples to split ( $min\_samples\_split$ ) = 2 or





264 5; minimum samples per leaf (`min_samples_leaf`) = 1 or 2; and number of features  
 265 considered at each split (`max_features`) = 'sqrt' or 0.5. Despite testing this range of  
 266 values, model performance was relatively stable, with test  $R^2$  scores varying modestly  
 267 across configurations. All models were implemented using scikit-learn ([https://scikit-](https://scikit-learn.org/stable/modules/generated/sklearn.ensemble.RandomForestRegressor.html)  
 268 [learn.org/stable/modules/generated/sklearn.ensemble.RandomForestRegressor.html](https://scikit-learn.org/stable/modules/generated/sklearn.ensemble.RandomForestRegressor.html)).  
 269 To validate the model, we randomly split our dataset (approximately 3000 counties in  
 270 the contiguous USA) into 5 equal subsets, known as *folds*. We then performed 5-fold  
 271 cross-validation, where each iteration trained the model on 4 of these folds and used  
 272 the remaining fold as a testing set to evaluate model performance. Each fold served  
 273 once as the testing set during the 5-fold cross-validation. The best-performing model  
 274 was selected based on the highest mean cross-validated  $R^2$  and used for further  
 275 interpretation, as our primary objective was to assess the relative importance of input  
 276 features.

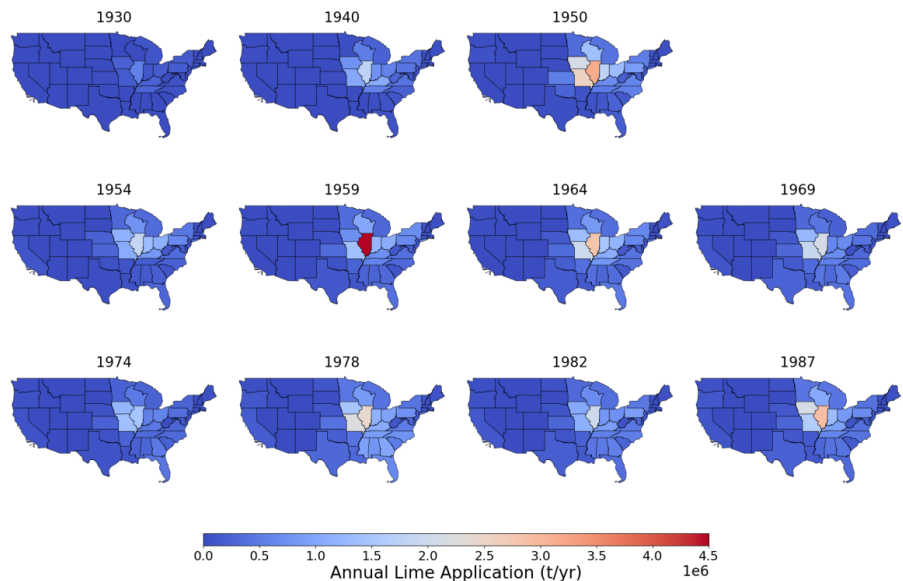
277 Random Forest models further provide an assessment of variable importance using  
 278 permutation importance, which quantifies the relative contribution of each predictor  
 279 variable by measuring the decrease in model performance when the variable's values  
 280 are randomly shuffled (Altmann et al. 2010). These scores were averaged across the  
 281 five folds to ensure a robust estimation of variable influence. In addition, partial  
 282 dependence analyses offer insights into the marginal effect of each predictor on the  
 283 target outcome, holding all other variables constant. This allows visualization of non-  
 284 linear relationships and interaction effects between predictors and target outcome. The  
 285 combination of linear (Pearson correlation) and non-linear (Random Forest model)  
 286 approaches provides a more comprehensive view of the environmental factors  
 287 influencing lime application, revealing the dominant drivers of the spatial variability  
 288 in lime application. We applied this statistical analysis to three response variables: (1)  
 289 annual lime applied (metric tons per year, normalized by hectare of land area), (2)  
 290 annual lime application rate (metric tons per hectare of limed area per year), and (3)  
 291 liming frequency (ratio of limed area to total cropland area per year) in each county.

292



293 **3. Results**

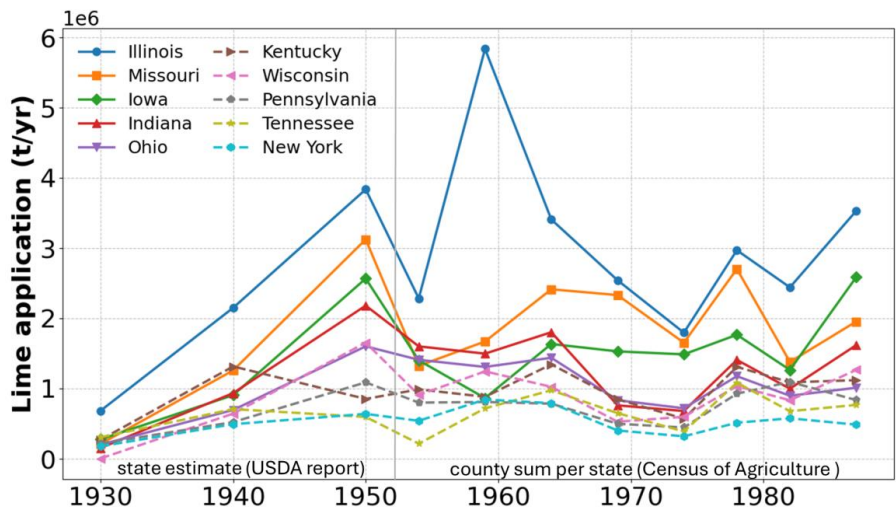
294 **3.1 Spatial-temporal pattern of lime application**



295

296 Figure 2 Annual lime application (metric tons per year, t/yr) per state in the contiguous U.S.  
297 from 1930 to 1987. The data from 1930 to 1950 is from the state-level data from the USDA  
298 report, while the data between 1954-1987 is summed from the county-level data from the  
299 Census reports. The data represents the condition during each specific year, not including the  
300 period in between.

301



302

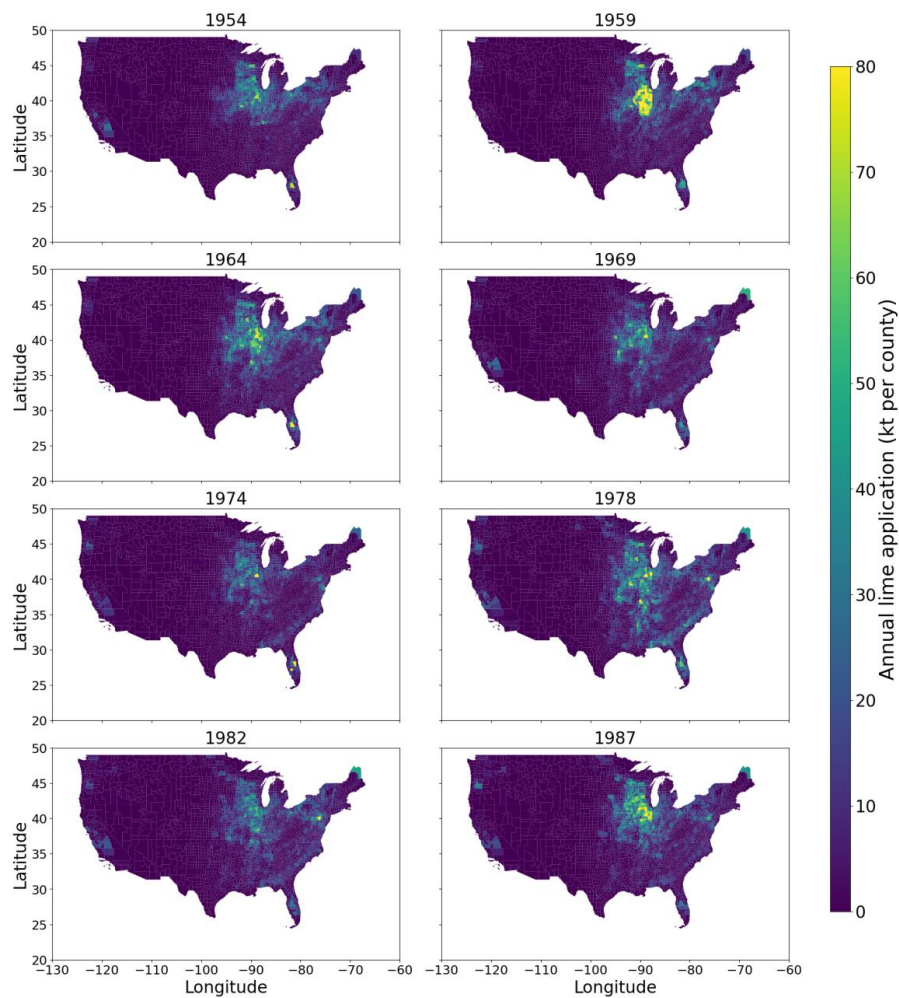
303 Figure 3 Temporal variations in lime applied (metric tons per year, t/yr) per state in the



contiguous U.S. from 1930 to 1987. The data from 1930 to 1950 is from the state-level data from the USDA report, while the data between 1954–1987 is summed from the county-level data from the Census reports. The figure shows the top 10 states ranked by state-level total lime application during 1954–1987, with Illinois applying the most lime, followed by Missouri, Iowa, Indiana, Ohio, Kentucky, Wisconsin, Pennsylvania, Tennessee, and Georgia.

The growth in state-level estimates of lime use from 1930–1987 was disproportionately concentrated in the Midwest (Fig. 1). A rapid increase in application occurred between 1930 and 1950, likely corresponding to the onset of the Green Revolution in the 1940s, which introduced synthetic fertilizers and modern agricultural technologies (Smil, 2004). The increasing trend does not persist after 1950, and the total amount of lime applied exhibits considerable temporal variation (Fig. 2). In the early 1950s, most of the lime was applied in the Midwest states. In 1959 an abrupt increase of lime is shown in Illinois (exceeding 6 Mt lime year<sup>-1</sup>), which could potentially be due to a combination of soil management needs, and government programs or educational initiatives (such as the Illinois Voluntary Limestone Program, <https://www.iaap-aggregates.org/agricultural-lime.html>). Starting from the 1960s, there is a gradual increase of lime application extending into the southern U.S. and particularly in the southeastern coast, reaching a peak in 1978, which Georgia alone exceeded 1 Mt lime year<sup>-1</sup> statewide (Fig. 2). In the 1980s lime application slightly declined on the southeastern coast but maintained a similar spatial pattern to the 1960s. Fig. 3 & Fig. 4 show the county-level lime application mass from 1954 to 1987, highlighting lime application being initially concentrated around the Midwest in the 1950s, a gradual decline in the later 60s to early 70s, an increase and expansion to the South and Southeast in the late 70s, and a slight decline in the 80s.

328

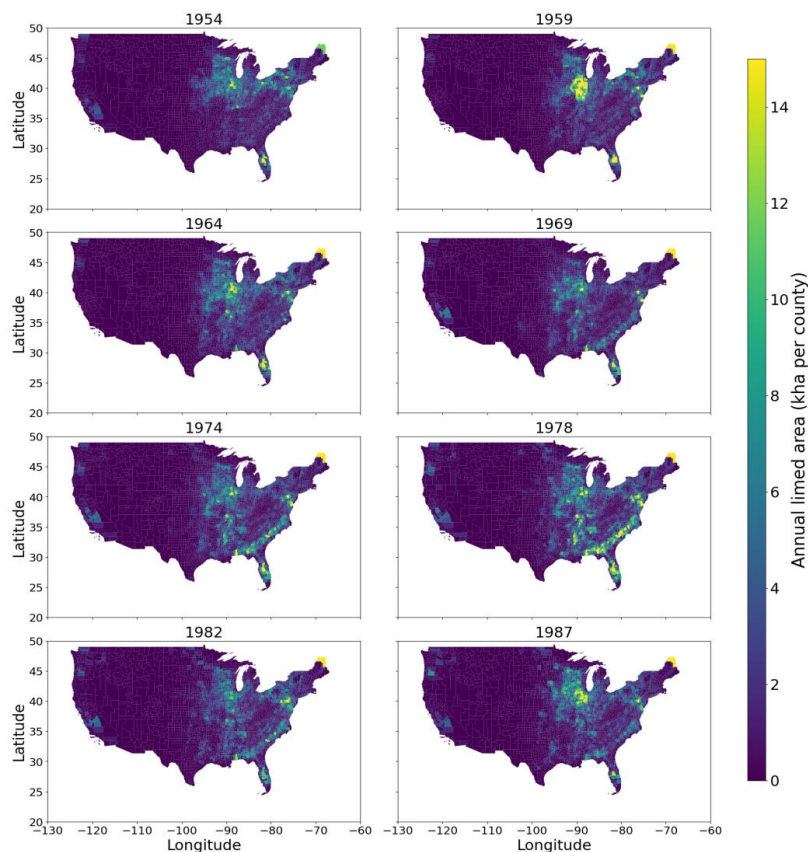


329  
330 Figure 4 Annual lime application (kilotons, kt) per county in the contiguous U.S. for the eight  
331 census years from 1954 to 1987, as reported by the Census Bureau. The maps display the lime  
332 application across counties with missing data temporarily interpolated and spatially filled, as  
333 described in the Methods section. The data only represents the condition during each specific  
334 census year, not including the period in between.

335  
336



337 The temporal-spatial pattern of county-level limed area (Fig. 5) approximately  
338 resembles that of limed applied mass, which the counties with the higher area of lime  
339 application are also distributed in the midwestern regions. Limed areas were high in  
340 the Midwest by the 1950s and gradually extended southward into the eastern and  
341 southeastern coasts starting from the 1970s. However, compared to limed weight, the  
342 limed area in the southeastern coast shows a more significant signal, suggesting that  
343 an equivalent amount of lime is applied over a larger area in the southeast. Compared  
344 to the states with the highest lime weight applied being all in the Midwestern regions,  
345 the states that have the highest limed area include both midwestern and southeastern  
346 states (Fig. S3). The relationship between limed weight and limed area across regions  
347 reflects the variation of lime application rates across regions, which may be  
348 influenced by local soil and climate properties (Fig. S4) .

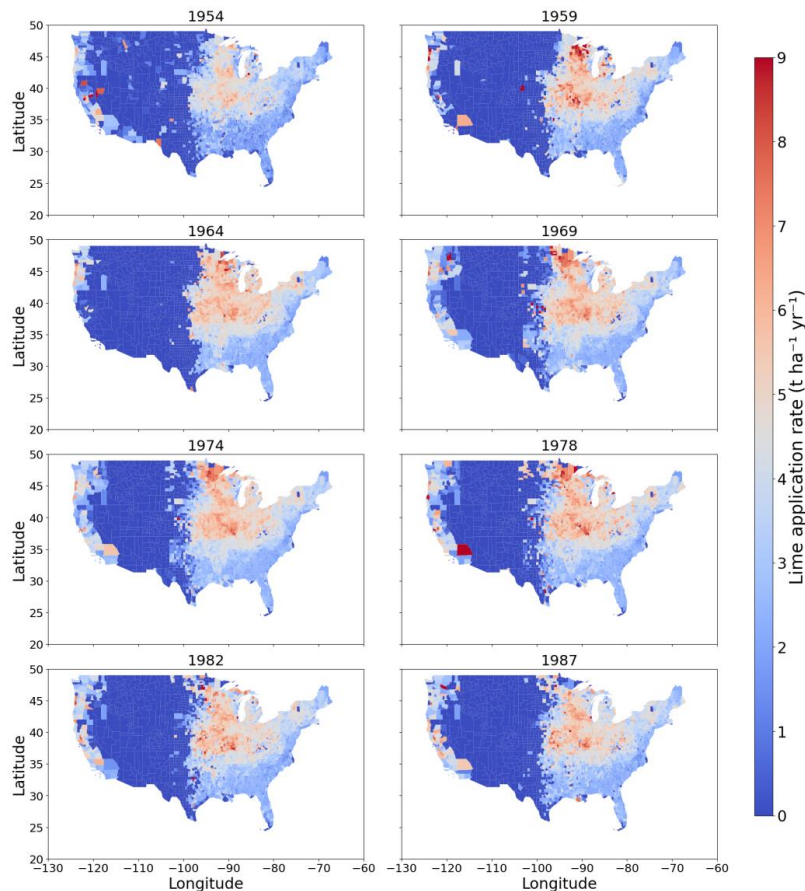


349  
350 Figure 5 Annual limed area (kilohectares, kha) per county in the contiguous U.S. for eight  
351 census years from 1954 to 1987, as reported by the Census Bureau. The maps display lime  
352 application across counties, with missing data temporarily interpolated and spatially filled, as  
353 described in the Methods section.

354



355 The lime application rate for each county (Fig. 6), calculated as the ratio of applied  
 356 mass to area of lime application, shows a sharp transition between the western and  
 357 eastern parts of the U.S. Most of the lime was applied only in the eastern half of the  
 358 U.S., while only a few coastal regions received lime in the western half of the US.  
 359 Lime application rates were relatively lower during the 1950s but reached a similar  
 360 level from the 1960s to the 1980s. Fig. S4 shows that the midwestern states of the  
 361 U.S. had the highest lime application rate ( $> 4 \text{ t ha}^{-1} \text{ yr}^{-1}$ ). Notably, Michigan and  
 362 Minnesota, though not ranked as the top 10 states of total amount applied (Fig. 2),  
 363 have a high lime application rate ranking (8<sup>th</sup> and 10<sup>th</sup> respectively). In comparison  
 364 with the mid-west region, the eastern, southeastern, and southern regions of the U.S.,  
 365 have lower lime application rates ( $\sim 2 \text{ t ha}^{-1} \text{ yr}^{-1}$ ).



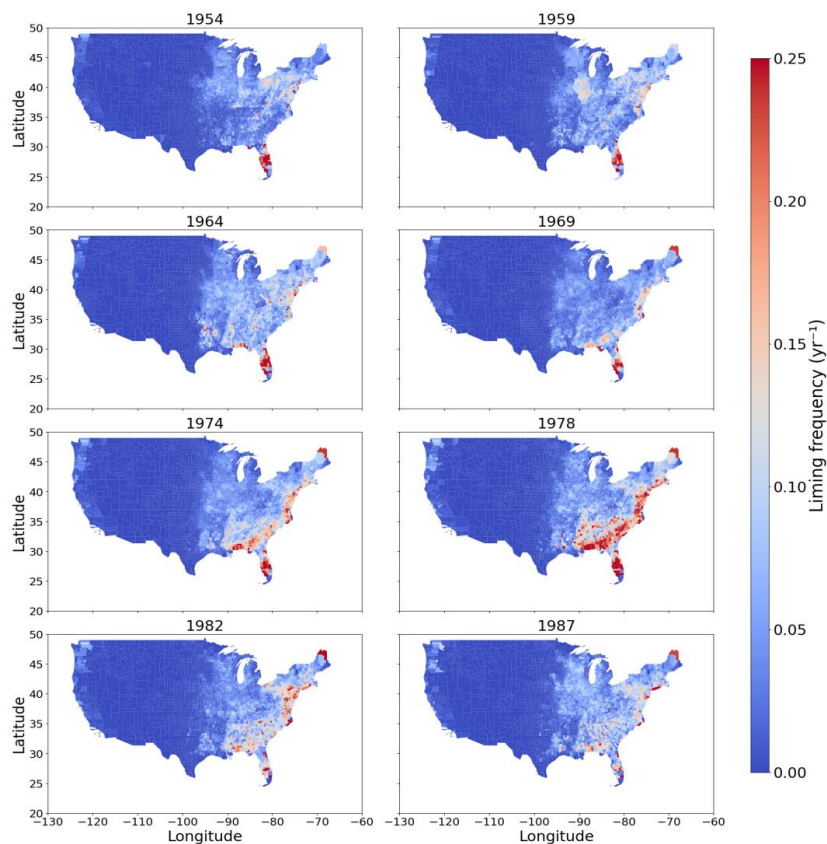
366  
 367 Figure 6. Lime application rate ( $\text{t ha}^{-1} \text{ yr}^{-1}$ ) for each county in the contiguous U.S. from 1954  
 368 to 1987. Rates were calculated as the total lime applied (t) divided by the limed area (ha)  
 369 for each census year. The eight maps represent the eight census years that reported data during  
 370 this period.

371





372 The liming frequency, calculated as the ratio of agricultural land limed to total  
 373 agricultural land each year, reveals a distinct Midwest–Southeast contrast in the  
 374 eastern half of the US. (Fig. 7). The Southeast generally shows a higher frequency  
 375 compared to the Midwest. In the 1950s, most regions had a low ratio ( $<10\%$  of  
 376 agricultural lands limed per year), with only Florida and a few areas along the eastern  
 377 seaboard exhibiting higher values ( $\sim 20\%$ ). A marked increase in this ratio occurred  
 378 throughout the Southeast beginning in the 1960s, peaking at  $\sim 30\%$  in 1970. In  
 379 contrast, the Midwest consistently maintained much lower values, with only  $\sim 5\%$  of  
 380 cropland limed annually. Assuming uniform application across cropland, this  
 381 corresponds to an average liming interval of once every 20 years in the Midwest (Oh  
 382 & Raymond, 2006), versus once every 5 years in the Southeast. This contrasting  
 383 spatial pattern in both Fig. 6 (lime application rate) and Fig. 7 (liming frequency)  
 384 highlights the spatial differences in lime management, likely driven by regional  
 385 variations in climate and soil properties.



386  
 387 Figure 7. Liming frequency ( $\text{yr}^{-1}$ ) for each county in the contiguous U.S. from 1954 to 1987,  
 388 calculated as the ratio of limed area to total cropland area. The eight maps represent the eight  
 389 census years, showing the fraction of cropland treated with lime annually.

390





### 3.2 Statistical analysis of environmental controls

We examined potential environmental predictors of lime-application patterns in terms of three separate outcome variables: (a) total lime applied per county, (b) lime application rate, and (c) liming frequency. After removing the highly correlated predictors from the initial environmental parameters (Fig. S5, Table S2), our Random Forest models demonstrated strong explanatory power, with  $R^2$  values ranging from 0.83 to 0.86 on test data (20% of data) (Fig. 8), while the  $R^2$  on the training set (80% of data) averaged 0.97 to 0.98, indicating strong predictive performance with limited overfitting.

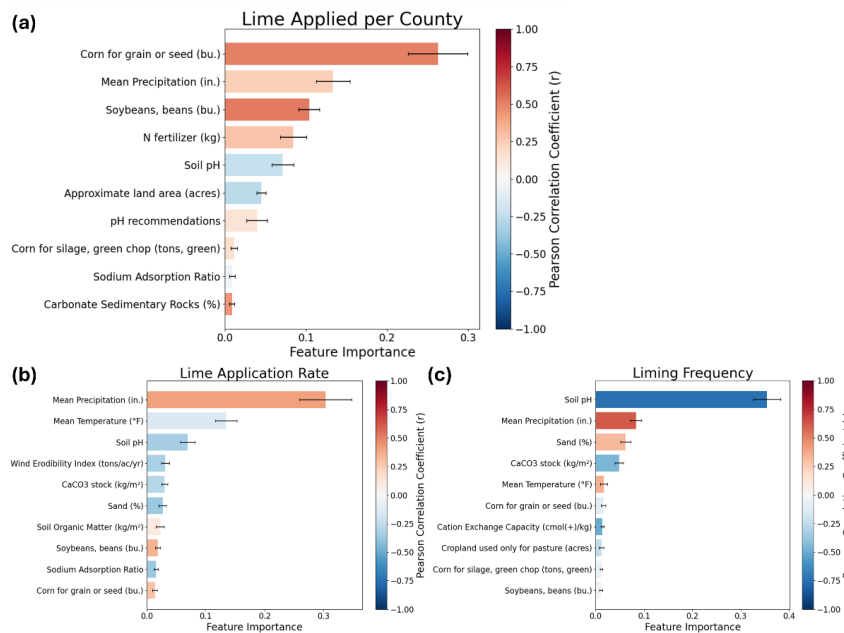


Figure 8 Statistical and machine learning analysis of average annual (a) total lime applied per county per year, normalized by county land area ( $\text{t ha}^{-1} \text{yr}^{-1}$ ), (b) lime application rate ( $\text{t ha}^{-1} \text{yr}^{-1}$ ), and (c) liming frequency ( $\text{yr}^{-1}$ ) per county across the three census years from 1978 to 1987. For (b) and (c) we ran the analysis only on the counties that have greater than 10% cropland by area, to minimize the influence of counties that have minimal agriculture. The bar length represents the permutation importance of all variables in the random forest model, with the error bar showing the standard error for the permutation importance across the 5-folds of cross validation training. Only the top ten variables with the highest permutation importance are shown here. The colors represent the Pearson correlation coefficients with the target variable.

For the total lime applied per county, the most influential predictors derived from the Random Forest model were corn grain production, mean precipitation, and soybean production, all of which showed strong positive associations. These were



415 followed by N fertilizer use, soil pH, approximate land area, and pH  
 416 recommendations. Partial dependence plots (Fig. S6) indicate that counties with  
 417 greater crop production, higher precipitation, lower soil pH elevated fertilizer inputs,  
 418 and higher pH targets tend to apply more lime. With respect to the lime application  
 419 rate, the most important variable was mean precipitation, positively correlated with  
 420 the response, followed by mean temperature, soil pH, wind erodibility index, and  
 421 carbonate mineral stocks, all showing negative correlations. Additional predictors  
 422 included sand percentage (negative correlation), soil organic matter (positive  
 423 correlation) (Fig. 8b). Partial dependence analyses reveal increased lime application  
 424 rates in wetter and colder regions with lower soil pH, wind erodibility, carbonate  
 425 mineral stocks, sand percentage, and higher soil organic matter (Fig. S7). For liming  
 426 frequency, soil pH emerged as the dominant predictor, exhibiting a strong negative  
 427 correlation. Other important predictors included mean precipitation and sand  
 428 percentage (positive correlations), and carbonate mineral stocks, mean temperature  
 429 (negative correlations). Counties with lower soil pH, lower carbonate minerals, higher  
 430 sand content, and greater rainfall exhibited more frequent liming practices (Fig. 8c,  
 431 Fig. S8). It is worth noting that the relative ranking of these features may change  
 432 slightly with different hyperparameter settings in the RF model, the important features  
 433 in the model reveal important information of the underlying relationships. Overall, the  
 434 average quantity of lime applied in counties is largely influenced by agricultural  
 435 management practices, including crop production, fertilizer application, and soil pH  
 436 recommendations, as well as climate. In contrast, the application rate and frequency  
 437 are predominantly governed by regional climatic conditions and soil properties.

438



## 439 4. Discussion

### 440 4.1 Spatial and temporal change of county-level lime application

441 From our results we showed that most agricultural lime was applied in the Midwest  
 442 region of the continental U.S. during the years 1930-1987. This pattern likely reflects  
 443 higher precipitation and lower soil pH in the east compared to western U.S.  
 444 (Hoffmeister, 1947; Wieczorek, 2019). Precipitation leaches base cations (calcium,  
 445 magnesium) and depletes buffering agents such as  $\text{CaCO}_3$ , causing soil pH to drop  
 446 nonlinearly as rainfall increases (Slessarev et al. 2016). Consistent with this  
 447 observation, we found that the average soil pH (from SSURGO) has a strong negative  
 448 correlation with average precipitation (from PRISM) (Pearson's  $r=-0.80$ ; Fig. S5).

449 An extensive amount of lime is applied in the Corn Belt in the American Midwest,  
 450 which has more than 70 million ha in corn and soybean rotation and is one of the most  
 451 intensively managed agricultural regions globally (Green et al., 2018). Corn grain  
 452 production emerged as the strongest predictor of total lime applied per county in the  
 453 permutation-based model (Fig. 8a), followed by mean precipitation, soybean  
 454 production, nitrogen fertilizer use, and soil pH. We further show that there is a  
 455 gradual expansion of the lime application area (Fig. 5), correlating with the  
 456 expansion, in particular a southward extension, of corn and soybean production area  
 457 (Fig. S9, S10), as there was a slight decline in tobacco and cotton production (Fig.  
 458 S13, S14). There is also an increase in lime application on the southeastern coast  
 459 starting from the 1960s and 1970s, matching the increase of agriculture in the region.  
 460 However, lime application along the southeastern coast declined in the 1980s,  
 461 potentially due to a decrease of agricultural land-use resulting from urbanization and  
 462 conservation programs (Napton et al., 2010). Additionally, the growth of the regional  
 463 lumber industry during this period may have further reduced agricultural acreage,  
 464 contributing to the observed decline in lime application (Prestemon & Abt, 2002).

465 The higher importance of corn and soybeans in determining the amount of lime  
 466 applied compared to other crop types (e.g., hay, wheat, cotton, tobacco) (S11-S15),  
 467 may be primarily related to the intensity of associated farming practices. Corn and  
 468 soybeans constitute a significantly larger portion of the US's harvested grains (Zulauf  
 469 et al., 2023) and are often grown with higher intensity (Annan et al., 2024). The  
 470 intensive agriculture, including frequent fertilizer inputs, acidifies the soil,  
 471 necessitating lime application. Additionally, soybeans are also a nitrogen fixing  
 472 species, which can promote nitrification and increase soil acidification (Nyantsanga &  
 473 Pierre, 1973). Although other crops like alfalfa may be more pH-sensitive, their  
 474 production is less fertilizer intensive and may result in a weaker apparent relationship  
 475 with lime application (Mallarino et al., 2011). As for non-alfalfa hay systems,  
 476 relatively low profit margins may deter investment in fertilizer and lime applications.



477 Furthermore, counties in the Midwest that predominantly grow corn and soybeans  
 478 tend to have a higher proportion of cropland relative to total land area (Fig. S16),  
 479 which adds to the association between production of these crops and the amount of  
 480 lime applied relative to total county area. Taken together, these results suggest that  
 481 while broad patterns such as the association between lime use and corn-soybean  
 482 agriculture are evident, the drivers of lime application are complex, regionally  
 483 dynamic, and shaped by interacting environmental, agronomic, and economic factors.  
 484 Our machine learning framework highlights just one of the perspectives to these  
 485 complex relationships, which we have shown is more dynamic at the regional scale  
 486 than commonly recognized.

#### 487 **4.2 Spatial distribution of lime application rate and frequency**

488 We show a spatial pattern of higher liming application rates ( $> 4 \text{ t ha}^{-1} \text{ yr}^{-1}$ ) and  
 489 longer liming intervals ( $\sim 20$  years) in the Midwest and lower application rates with  
 490 shorter intervals ( $\sim 5$  years) near the southeastern coastal regions (Fig. 6-7).  
 491 Permutation importance analysis (Fig. 8b) shows that mean precipitation is the most  
 492 important predictor of lime application rate, followed by mean temperature, soil pH,  
 493 wind erodibility index, carbonate mineral stock, sand content, and soil organic matter.  
 494 In contrast, liming frequency is most strongly explained by soil pH, followed by mean  
 495 precipitation, sand content, and carbonate mineral stock (Fig. 8c). Together, these  
 496 findings suggest that lime application practices are governed by a complex interplay  
 497 between acidification drivers and soil buffering responses.

498 The required lime application rate and liming frequency is heavily dependent on the  
 499 soil acidification rate and pH buffering capacity (Xu et al., 2022; Kanzaki et al. 2024).  
 500 Higher precipitation leads to faster leaching of base cations, while regions of warmer  
 501 temperatures lead to faster decomposition and organic acid release, with both  
 502 processes leading to faster soil acidification (Ulrich, 1986). Both processes promote  
 503 faster soil acidification. However, precipitation appears to be a necessary condition  
 504 for acidification to manifest at scale, while temperature acts both as a direct driver and  
 505 an indirect proxy for soil characteristics. Specifically, warmer regions tend to have  
 506 more weathered soils with low buffering capacity, while cooler regions, particularly  
 507 in the Midwest, often retain glacially derived carbonates and clay minerals that confer  
 508 greater buffering potential. This helps explain the somewhat counterintuitive negative  
 509 correlation between temperature and application rate: although warmer regions  
 510 acidify faster, their low-buffering soils allow for smaller lime doses per application,  
 511 whereas cooler, more buffered soils require larger doses when they are eventually  
 512 limed. This pattern is further supported by the partial dependence plots (Fig. S7),  
 513 which reveal a distinct threshold-like response: lime application rate increases steeply  
 514 with precipitation beyond approximately 750–800 mm/year, and declines sharply with



515 temperature above 12–13°C. These non-linear relationships imply that precipitation is  
 516 a key threshold variable enabling acidification, while temperature's response reflects  
 517 both its role in driving acid production and its spatial correlation with regional  
 518 differences in soil buffering capacity.

### 519 **4.3 Regional variation in buffering capacity and liming strategy**

520 Resisting the higher acidification rates associated with warmer, wetter conditions in  
 521 the southeastern USA likely entail more frequent lime additions. However, these  
 522 regions typically do not require higher application rates per event, as their soils  
 523 exhibit relatively low pH buffering capacity. The pH buffering capacity of soils varies  
 524 systematically between the Midwest and the Southeast, which is another main cause  
 525 in the spatial patterns in lime application rate and frequency. Soil buffering capacity is  
 526 influenced by naturally occurring carbonate minerals ( $\text{CaCO}_3$  and  $\text{CaMg}(\text{CO}_3)_2$ ),  
 527 which can directly neutralize acidity when it dissolves. The influence of carbonate  
 528 minerals is particularly relevant in some Midwestern soils, where residual carbonate  
 529 from glacial parent materials helps maintain higher soil pH values. In contrast,  
 530 Southeastern soil typically lacks carbonate minerals, making them more vulnerable to  
 531 acidification (Fig. S17). In addition to carbonates, cation exchange capacity (CEC)  
 532 helps to resist swings in pH by capturing or releasing adsorbed base cations and  
 533 protons (Brady & Weil, 2016). CEC also varies systematically between the  
 534 southeastern and midwestern USA. Several factors contribute to the lower CEC  
 535 observed in highly-weathered southeastern soils (Fig. S18), which are dominated by  
 536 low activity clay minerals, coarser particle sizes, elevated concentrations of detrital  
 537 quartz (Woodruff et al., 2009), and lower organic matter content (Solly et al., 2020).  
 538 Conversely, soils in the upper Midwest are derived from glacial parent materials and  
 539 loess (Borker et al. 2018), which are often characterized by higher organic matter  
 540 contents, smaller soil particle sizes, and higher activity clay minerals (Fig. S19, S20),  
 541 providing more CEC and better pH buffering capacity. These finer-textured soils are  
 542 also generally less erodible by wind, consistent with the observed negative correlation  
 543 between wind erodibility and lime application rate.

544 Although CEC and carbonate content are key conceptual components of buffering  
 545 capacity, they did not emerge as top predictors in the permutation importance  
 546 analysis. This is likely due to collinearity with soil pH, which reflects the integrated  
 547 outcome of both acidification pressure and buffering history. In this sense, soil pH  
 548 serves as a practical proxy for underlying soil properties that govern liming  
 549 frequency. The spatial correlation is particularly relevant in the southeastern U.S.,  
 550 where low soil pH tends to co-occur with both low CEC and minimal carbonate  
 551 content, rendering pH a strong indicator of inherently low buffering capacity. A  
 552 similar rationale applies temperature's high variable importance in predicting lime



553 application rate, which as discussed earlier, is likely to reflect its spatial co-correlation  
 554 with the degree of soil weathering and associated buffering capacity.

555 In summary, both climatic drivers (temperature precipitation) and soil mineralogy  
 556 (including soil pH, carbonate presence, wind erodibility, soil organic matter, and  
 557 particle size distribution) together explain the spatial variation of lime application rate  
 558 and frequency. The Midwestern region requires less frequent but larger amounts of  
 559 lime application to maintain soil pH, while the southeastern region requires more  
 560 frequent but smaller amounts of lime application to effectively maintain optimal pH  
 561 levels (Ross et al., 1964).

#### 562 **4.4 Temporal variability of total agricultural lime application.**

563 We further hypothesize that economics may have played an important role in  
 564 shaping the temporal variability of lime use. As shown in Fig. S21, the late 1960s and  
 565 early 1970s witnessed a surge in fertilizer expenditures and a drop in fertilizer cost,  
 566 likely driven by technological advances, leading farmers to prioritize fertilizer over  
 567 liming. This behavior may have contributed to the notable decline in lime application  
 568 during this period. The immediate yield gains from increased fertilizer use could have  
 569 overshadowed the longer-term implications of reduced liming, particularly given that  
 570 soil acidification tends to accumulate gradually over multi-year or decadal  
 571 timescales. In 1974, fertilizer prices rebounded sharply in conjunction with the global  
 572 fuel crisis (Brunelle et al., 2015), aligning with the most pronounced dip in lime  
 573 application rate observed that year. In the late 1970s to early 1980s, farm incomes  
 574 peaked, and input costs stabilized (Fig. S22), creating conditions conducive to greater  
 575 focus on long-term soil health. Consequently, this period saw a resurgence in lime  
 576 application. It is also likely that this period marked growing recognition of the soil  
 577 acidification problem, as several studies on liming and soil pH management were  
 578 published during this time (Nyborg & Hoyt, 1978; Bache, 1980). By the late 1980s,  
 579 declining crop sales and lower input costs suggest a period of adjustment and input  
 580 optimization, potentially explaining the modest reduction in total lime applied (Fig.  
 581 S22)

#### 582 **4.5 Comparison of total lime application estimate with West & McBride (2005)**

583 Fig. 9 shows a comparison between the total lime application in our dataset and the  
 584 national estimate of lime application in West & McBride (2005), which to our  
 585 knowledge is the first national estimate of agriculture lime applied in the US. Our  
 586 study and West & McBride both show that lime application increases very slowly  
 587 starting from the 1910s but started to rapidly increase in ~1930 and reach a high range  
 588 of values, exceeding 20 Mt/yr around 1950. However, there are some discrepancies  
 589 from 1950 through 1980. West & McBride estimated the total lime applied to  
 590 continue to increase, particularly during the 1970s, which exceeded 30 Mt per year,  
 591 while our dataset shows the total lime applied to be more consistent in the range of



15-25 Mt per year. Oh & Raymond (2006) conducted a regional study based on the county-level liming dataset from the Census reports and first noticed disagreements in the trends of the regional sum of the county-level data and the national estimate from West & McBride. However, Oh and Raymond's study was done over a small region of the US, and a spatially explicit dataset of lime application across the U.S. was not yet available at that time to address the uncertainty in the total lime estimates.

Here we revisited the discrepancy between the national estimate of lime application and that of West & McBride (2005). We suggest that the difference is primarily due to uncertainties in the methodology of West & McBride, which relies on data from the Mineral Yearbook (U.S. Bureau of Mines, 1927–1996; U.S. Geological Survey, 1906–1927, 1997–2002). The Mineral Yearbook classifies crushed stones by their intended use, with agricultural limestone being a sub-category of “agricultural uses”. We noticed roughly half of crushed stones are classified under an “unspecified end-use” category. West & McBride estimated total agricultural lime by combining explicitly labeled agricultural limestone with an inferred fraction (approximately 2–3%) from the unspecified category, assuming proportional distribution across all categories.

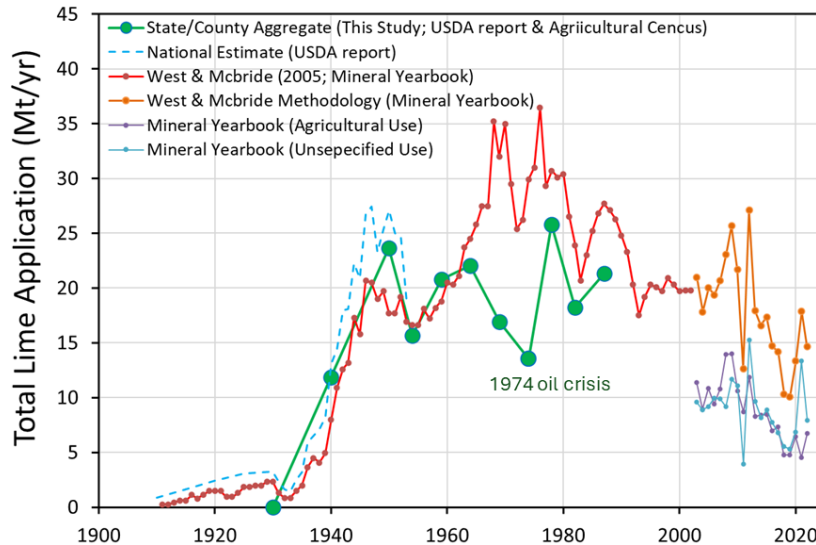


Figure 9 Comparison of national estimates of the total weight of lime applied (tons) over time. Our study's compilation (state-level aggregate (1930–1950) from the USDA report and county-level aggregate (1954–1987) from the Agriculture Census) are shown as green dots. The green dashed line represents the national estimate reported annually in the USDA report (1910–1950). The red line represents national estimates from West & McBride (2005). The orange dots indicate the extended total lime applied calculation based on the Mineral Yearbook using methods from West & McBride (2005). The purple line represents the fraction of lime applied for agricultural use, and the blue line reflects the unspecified end-use fraction, scaled by the ratio of agricultural use to total specified end-use, representing the





619 potential lime used for agriculture.

620

621 We further extended the methodology of West & McBride (2005) until 2022  
 622 (orange line) (data extracted from [https://www.usgs.gov/centers/national-minerals-](https://www.usgs.gov/centers/national-minerals-information-center/crushed-stone-statistics-and-information)  
 623 [information-center/crushed-stone-statistics-and-information](https://www.usgs.gov/centers/national-minerals-information-center/crushed-stone-statistics-and-information)). We aggregated the  
 624 number of crushed stones specified as “agricultural limestone” with an estimated  
 625 percentage of agricultural lime within the “unspecified” end-use category. We show  
 626 that the fraction of data in the unspecified end-use category (blue line; assumed to be  
 627 used for agriculture) is similar in magnitude as those specified to be used for  
 628 agriculture (purple line). This suggests that ~50% of the estimate in West & McBride  
 629 is sensitive to how “unspecified” use is interpreted, which leads to high uncertainty  
 630 and potential biases in total estimate. Given that agricultural limestone is such a small  
 631 fraction of the total unspecified crushed rocks (2–3%), small variations in this  
 632 percentage would lead to a large impact on the total estimated agricultural lime.  
 633 Therefore, we believe that our compiled dataset here offers a better representation of  
 634 the total amount of lime applied in the US, and that 15–25 Tg per year could be a  
 635 more accurate range instead of the 25–35 Tg per year suggested in West & McBride.

636 Based on our extended analysis with West & McBride (2005)’s methodology, we  
 637 also show that the total annual applied lime in the last two decades varied between  
 638 10–25 Tg per year, but did not surpass the peak in the 1970s. A similar trend is shown  
 639 in the total nutrient input via fertilizers, where there is no significant increase of N and  
 640 P input since 1980 (Fig. S23). This suggests that the amount of agricultural lime  
 641 applied likely did not increase since then, providing evidence that the temporal range  
 642 of our dataset in this study covered the period when lime application reached its full  
 643 extent in the USA.

## 644 5. Data Availability

645 The dataset presented in this study will be made publicly available on Zenodo at  
 646 <https://doi.org/10.5281/zenodo.15758275> (Tsao, S. S.-E., Surhoff, T. J., Amatulli, G.,  
 647 & Raymond, P. (2025). Agricultural lime application across the contiguous United  
 648 States, 1930–1987 [Data set]. Zenodo.). Reviewers can access the dataset during peer  
 649 review via the private preview link:  
 650 [https://zenodo.org/records/15758275?preview=1&token=eyJhbGciOiJIUzUxMiJ9.eyJpZCI6IjdiN2VkNDkwLTE1YjctNGNiZC1iMjY2LTBlOGMzZjkzYTZmZSIsImRhdiGEiOnt9LCJyYW5kb20iOiJjZTFmNTA4ZDliZGU4NDQzOGNIMDkzMtYxNjJkMDEQxNSJ9.2yChJg8txya\\_OZzIo6nmgFAjVj-haakBfMtFcLrDukLXaZxKVM19i-DrUZ3kKuI443l4z1KjREqIKiOIHbJ42g](https://zenodo.org/records/15758275?preview=1&token=eyJhbGciOiJIUzUxMiJ9.eyJpZCI6IjdiN2VkNDkwLTE1YjctNGNiZC1iMjY2LTBlOGMzZjkzYTZmZSIsImRhdiGEiOnt9LCJyYW5kb20iOiJjZTFmNTA4ZDliZGU4NDQzOGNIMDkzMtYxNjJkMDEQxNSJ9.2yChJg8txya_OZzIo6nmgFAjVj-haakBfMtFcLrDukLXaZxKVM19i-DrUZ3kKuI443l4z1KjREqIKiOIHbJ42g)  
 651  
 652  
 653  
 654

655 This dataset provides spatially explicit estimates of agricultural lime application  
 656 across the contiguous United States, including: 1. State-level data from 1930 to 1950,  
 657 and 2. County-level data from 1954 to 1987.

658 Key variables include total lime applied (metric tons), area limed (hectares),



659 application rate ( $\text{t ha}^{-1} \text{yr}^{-1}$ ), liming intensity ( $\text{t ha}^{-1} \text{yr}^{-1}$ ), and liming frequency ( $\text{yr}^{-1}$ ),  
 660 which are visualized in Figures 4–7.

661 The county-level shapefile contains the following columns:

- 662     -Year: Year of observation
- 663     -GEOID: Unique identifier for each U.S. county
- 664     -LimeTons: Total lime applied (metric tons)
- 665     -LimeArea: Area limed (hectares)
- 666     -CropArea: Total cropland area (hectares)
- 667     -LimeRate: Lime application rate ( $\text{t ha}^{-1} \text{yr}^{-1}$ )
- 668     -LimeIntsty: Liming intensity ( $\text{t ha}^{-1} \text{yr}^{-1}$ )
- 669     -LimeFreq: Liming frequency ( $\text{yr}^{-1}$ )

670 All files are provided in open formats and are accompanied by metadata and  
 671 documentation to ensure transparency and reproducibility.

## 672 **6. Code Availability**

673 The code used for data processing, interpolation, and machine learning analysis will  
 674 be made publicly available upon publication of the final version of this manuscript. A  
 675 link to the repository will be provided at that time to ensure full reproducibility.

## 676 **7. Conclusion**

677 In this study, we compiled and presented the first spatially explicit data set of  
 678 agricultural lime application in the U.S. from 1930–1987. We showed that the  
 679 Midwest region has the most amount of lime applied, while the south and  
 680 southeastern coastal region of the U.S. has gradually increased lime application. We  
 681 also showed that the Midwest has a much longer liming interval and higher lime  
 682 application rate than the Southeastern and eastern US, while the liming frequency is  
 683 higher in the Southeastern and eastern regions than in the Midwest. Using machine  
 684 learning algorithms, we showed that on the county level, the total amount of lime  
 685 applied is largely influenced by the agricultural management practices (soybean and  
 686 corn production, fertilizers, soil pH recommendation) alongside natural factors such  
 687 as soil pH and climate. In contrast lime application rate and frequency are  
 688 predominantly governed by natural regional climate (e.g., precipitation, temperature)  
 689 and soil characteristics (e.g., soil pH, carbonate minerals, particle size distribution,  
 690 soil texture, organic matter). We further show that the temporal variability in total  
 691 lime application may be influenced by economic factors, including competing input  
 692 decisions such as fertilizer use, as well as fluctuations in fuel prices and farm income  
 693 and expenditure.

694 Our study synthesizes historical lime application practices across both temporal and



695 spatial dimensions, offering a critical observational baseline for future modeling  
 696 efforts. As liming gains attention for its potential role in carbon removal and climate  
 697 mitigation, understanding historical application patterns is essential for accurately  
 698 assessing its long-term carbon sequestration potential and contribution to greenhouse  
 699 gas reductions. Future research should build on these trends by integrating spatial and  
 700 temporal dynamics with variations in soil types, climate conditions, and other  
 701 environmental factors to comprehensively assess the viability of liming as a nature-  
 702 based climate solution.

703

## 704 **8. Author Contributions**

705 Conceptualization: MA, TJS, GA, CTR, NJP, PAR  
 706 Methodology: SST, TJS, GA, JG, EWS, CTR, SZ, PAR,  
 707 Software: GA  
 708 Validation: SST, TJS  
 709 Formal Analysis: SST, TJS, EWS  
 710 Investigation: SST, TJS, GA,  
 711 Resources: PAR  
 712 Data Curation: TJS, GA, SST  
 713 Writing – Original Draft: SST  
 714 Writing – Review & Editing: TJS, GA, BW, MA, EWS, JES, CTR, SZ, NJP, PAR  
 715 Visualization: SST  
 716 Supervision: PAR  
 717 Project Administration: PAR  
 718 Funding Acquisition: PAR

719

## 720 **9. Competing Interest**

721 The authors declare that they have no conflict of interest.

722

## 723 **10. Acknowledgements.**

724 10. This research was supported by the U.S. Department of Energy Earthshot  
 725 Initiative (grant no. DE-SC0024709). Additional support was provided by the Yale  
 726 Center for Natural Carbon Capture and the Yale School of the Environment. We  
 727 thank Rachel Sperling (Yale University Library) for assistance with data mining and  
 728 the Spatial Ecology summer workshop team for training and help in accelerating the  
 729 data-processing workflow. We are also grateful to all members of the Raymond Lab  
 730 and Saiers Lab for their constructive feedback during the preparation of this  
 731 manuscript.

732



## 733 References

- 734 Agegnehu, G., Amede, T., Erkossa, T., Yirga, C., Henry, C., Tyler, R., Nosworthy,  
 735 M. G., Beyene, S., and Sileshi, G. W.: Extent and management of acid soils for  
 736 sustainable crop production system in the tropical agroecosystems: A review, *Acta*  
 737 *Agric. Scand. Sect. B Soil Plant Sci.*, 71, 852–869,  
 738 <https://doi.org/10.1080/09064710.2021.1954239>, 2021.
- 739  
 740 Altmann, A., Toloşi, L., Sander, O., and Lengauer, T.: Permutation importance: a  
 741 corrected feature importance measure, *Bioinformatics*, 26, 1340–1347, 2010.
- 742 Annan, K., Fausti, S. W., Van der Sluis, E., and Kolady, D. E.: Corn Acreage  
 743 Intensification Levels in US Corn Belt States, *J. Agric. Appl. Econ.*, 56, 353–372,  
 744 2024.
- 745  
 746 Bache, B. W.: The acidification of soils, in: *Effects of acid precipitation on terrestrial*  
 747 *ecosystems*, edited by: Dochinger, L. S. and Seliga, T. A., Springer US, Boston, MA,  
 748 183–202, [https://doi.org/10.1007/978-1-4613-3083-1\\_11](https://doi.org/10.1007/978-1-4613-3083-1_11), 1980.
- 749  
 750 Beerling, D. J., Kantzas, E. P., Lomas, M. R., Wade, P., Eufrasio, R. M., Renforth, P.,  
 751 Sarkar, B., Andrews, M. G., James, R. H., Pearce, C. R., Mercure, J.-F., Pollitt, H.,  
 752 Holden, P. B., Edwards, N. R., Khanna, M., Koh, L., Quegan, S., Pidgeon, N. F.,  
 753 Janssens, I. A., and Banwart, S. A.: Potential for large-scale CO<sub>2</sub> removal via  
 754 enhanced rock weathering with croplands, *Nature*, 583,  
 755 <https://doi.org/10.1038/s41586-020-2448-9>, 2020.
- 756  
 757 Breiman, L.: Random forests, *Mach. Learn.*, 45, 5–32, 2001.
- 758  
 759 Brunelle, T., Dumas, P., Souty, F., Dorin, B., and Nadaud, F.: Evaluating the impact  
 760 of rising fertilizer prices on crop yields, *Agric. Econ.*, 46, 653–666, 2015.
- 761  
 762 Burdett, H. and Wellen, C.: Statistical and machine learning methods for crop yield  
 763 prediction in the context of precision agriculture, *Precis. Agric.*, 23, 1553–1574,  
 764 <https://doi.org/10.1007/s11119-022-09897-0>, 2022.
- 765  
 766 De Klein, C. et al.: Chapter 11 - N<sub>2</sub>O Emissions from managed Soils and CO<sub>2</sub>  
 767 Emissions From Lime and Urea Application, *IPCC Guidelines for Natl. Greenh. Gas*  
 768 *Invent.*, Vol. 4, 11.1–11.54, 2006.
- 769  
 770 Falcone, J. A.: Estimates of county-level nitrogen and phosphorus from fertilizer and  
 771 manure from 1950 through 2017 in the conterminous United States, USGS Open-File



- Report, <https://doi.org/10.3133/ofr20201153>, 2021.
- Fageria, N. K. and Baligar, V. C.: Ameliorating soil acidity of tropical Oxisols by liming for sustainable crop production, *Adv. Agron.*, 99, 345–399, [https://doi.org/10.1016/S0065-2113\(08\)00407-0](https://doi.org/10.1016/S0065-2113(08)00407-0), 2008.
- Goulding, K. W. T.: Soil acidification and the importance of liming agricultural soils with particular reference to the United Kingdom, *Soil Use Manage.*, 32, 390–399, <https://doi.org/10.1111/sum.12270>, 2016.
- Green, T. R., Kipka, H., David, O., and McMaster, G. S.: Where is the USA Corn Belt, and how is it changing?, *Sci. Total Environ.*, 618, 1613–1618, 2018.
- Guo, J. H., Liu, X. J., Zhang, Y., Shen, J. L., Han, W. X., Zhang, W. F., Christie, P., Goulding, K. W. T., Vitousek, P. M., and Zhang, F. S.: Significant acidification in major Chinese croplands, *Science*, 327, 1008–1010, <https://doi.org/10.1126/science.1182570>, 2010.
- Haines, M., Fishback, P., and Rhode, P.: United States Agriculture Data, 1840–2012, Inter-university Consortium for Political and Social Research, <https://doi.org/10.3886/ICPSR35206.v4>, 2018.
- Hamilton, S. K., Kurzman, A. L., Arango, C., Jin, L., and Robertson, G. P.: Evidence for carbon sequestration by agricultural liming, *Glob. Biogeochem. Cycles*, 21, <https://doi.org/10.1029/2006GB002738>, 2007.
- Han, T., Cai, A., Liu, K., Huang, J., Wang, B., Li, D., Qaswar, M., Feng, G., and Zhang, H.: The links between potassium availability and soil exchangeable calcium, magnesium, and aluminum are mediated by lime in acidic soil, *J. Soils Sediments*, 19, 1382–1392, <https://doi.org/10.1007/s11368-018-2145-6>, 2019.
- Hartmann, J. and Moosdorf, N.: The new global lithological map database GLiM: A representation of rock properties at the Earth surface, *Geochem. Geophys. Geosyst.*, 13, Q12004, <https://doi.org/10.1029/2012GC004370>, 2012.
- Hoffmeister, H. A.: Alkali problem of western United States, *Econ. Geogr.*, 23, 1–9, 1947.



- 810 Hooda, P. S. and Alloway, B. J.: The effect of liming on heavy metal concentrations  
 811 in wheat, carrots and spinach grown on previously sludge-applied soils, *J. Agric. Sci.*,  
 812 127, 289–294, <https://doi.org/10.1017/S0021859600078448>, 1996.
- 813
- 814 Jeong, J. H., Resop, J. P., Mueller, N. D., Fleisher, D. H., Yun, K., Butler, E. E.,  
 815 Timlin, D. J., Shim, K.-M., Gerber, J. S., Reddy, V. R., and Kim, S.-H.: Random  
 816 Forests for global and regional crop yield predictions, *PLoS ONE*, 11, e0156571,  
 817 <https://doi.org/10.1371/journal.pone.0156571>, 2016.
- 818
- 819 Johnston, J. F. W.: On the use of lime in agriculture, 1849.
- 820
- 821 Kanzaki, Y., Planavsky, N., Zhang, S., Jordan, J., Suhrhoff, T. J., and Reinhard, C. T.:  
 822 Soil cation storage as a key control on the timescales of carbon dioxide removal  
 823 through enhanced weathering, *Authorea Preprints*, 2024.
- 824
- 825 Khaliq, M. A., Tarin, M. W. K., Jingxia, G., Yanhui, C., and Guo, W.: Soil liming  
 826 effects on CH<sub>4</sub>, N<sub>2</sub>O emission and Cd, Pb accumulation in upland and paddy rice,  
 827 *Environ. Pollut.*, 248, 408–420, 2019.
- 828
- 829 Knapp, W. J. and Tipper, E. T.: The efficacy of enhancing carbonate weathering for  
 830 carbon dioxide sequestration, *Front. Clim.*, 4,  
 831 <https://doi.org/10.3389/fclim.2022.928215>, 2022.
- 832
- 833 Liu, Z., Dreybrodt, W., and Liu, H.: Atmospheric CO<sub>2</sub> sink: Silicate weathering or  
 834 carbonate weathering?, *Appl. Geochem.*, 26, S292–S294,  
 835 <https://doi.org/10.1016/j.apgeochem.2011.03.085>, 2011.
- 836
- 837 Mallarino, A., Pagani, A., and Sawyer, J.: Corn and soybean response to soil pH level  
 838 and liming, *Iowa State Univ. Ext.*,  
 839 <https://dr.lib.iastate.edu/handle/20.500.12876/43941>, 2011.
- 840
- 841 Mehring, A. L., Adams, J. R., and Jacob, K. D.: Statistics on fertilizers and liming  
 842 materials in the United States, *USDA Soil and Water Conservation Research Branch*,  
 843 191, 1957.
- 844
- 845 Napton, D. E., Auch, R. F., Headley, R., and Taylor, J. L.: Land changes and their  
 846 driving forces in the Southeastern United States, *Reg. Environ. Change*, 10, 37–53,  
 847 2010.



- 848 Nyatsanga, T. and Pierre, W. H.: Effect of nitrogen fixation by legumes on soil  
 849 acidity, *Agron. J.*, 65, 936–940, 1973.
- 850
- 851 Nyborg, M. and Hoyt, P. B.: Effects of soil acidity and liming on mineralization of  
 852 soil nitrogen, *Can. J. Soil Sci.*, 58, 331–338, 1978.
- 853
- 854 Oh, N. and Raymond, P. A.: Contribution of agricultural liming to riverine  
 855 bicarbonate export and CO<sub>2</sub> sequestration in the Ohio River basin, *Glob.*  
 856 *Biogeochem. Cycles*, 20, <https://doi.org/10.1029/2005GB002565>, 2006.
- 857
- 858 Pagani, A. and Mallarino, A. P.: Soil pH and crop grain yield as affected by the  
 859 source and rate of lime, *Soil Sci. Soc. Am. J.*, 76, 1877–1886,  
 860 <https://doi.org/10.2136/sssaj2012.0119>, 2012.
- 861
- 862 Pedregosa, F., Varoquaux, G., Gramfort, A., Michel, V., Thirion, B., Grisel, O.,  
 863 Blondel, M., Prettenhofer, P., Weiss, R., Dubourg, V., Vanderplas, J., Passos, A.,  
 864 Cournapeau, D., Brucher, M., Perrot, M., and Duchesnay, É.: Scikit-learn: Machine  
 865 learning in Python, *J. Mach. Learn. Res.*, 12, 2825–2830, 2011.
- 866
- 867 Plummer, L. N. and Wigley, T. M. L.: The dissolution of calcite in CO<sub>2</sub>-saturated  
 868 solutions at 25 °C and 1 atmosphere total pressure, *Geochim. Cosmochim. Acta*, 40,  
 869 191–202, 1976.
- 870
- 871 Prestemon, J. P. and Abt, R. C.: Southern forest resource assessment highlights: The  
 872 southern timber market to 2040, *J. For.*, 100, 16–22, 2002.
- 873
- 874 Robertson, G. P., Paul, E. A., and Harwood, R. R.: Greenhouse gases in intensive  
 875 agriculture: Contributions of individual gases to the radiative forcing of the  
 876 atmosphere, *Science*, 289, 1922–1925, <https://doi.org/10.1126/science.289.5486.1922>,  
 877 2000.
- 878
- 879 Ross, G. J., Lawton, K., and Ellis, B. G.: Lime requirement related to physical and  
 880 chemical properties of nine Michigan soils, *Soil Sci. Soc. Am. J.*, 28, 209–212, 1964.
- 881 Semhi, K., Amiotte Suchet, P., Clauer, N., and Probst, J.-L.: Impact of nitrogen  
 882 fertilizers on the natural weathering-erosion processes and fluvial transport in the  
 883 Garonne basin, *Appl. Geochem.*, 15, 865–878, [https://doi.org/10.1016/S0883-](https://doi.org/10.1016/S0883-2927(99)00076-1)  
 884 [2927\(99\)00076-1](https://doi.org/10.1016/S0883-2927(99)00076-1), 2000.
- 885
- 886 Shaaban, M., Wu, Y., Wu, L., Hu, R., Younas, A., Nunez-Delgado, A., Xu, P., Sun,





- 887 Z., Lin, S., Xu, X., and Jiang, Y.: The effects of pH change through liming on soil  
 888 N<sub>2</sub>O emissions, *Processes*, 8, 702, <https://doi.org/10.3390/pr8060702>, 2020.
- 889 Slessarev, E. W., Lin, Y., Bingham, N. L., Johnson, J. E., Dai, Y., Schimel, J. P., and  
 890 Chadwick, O. A.: Water balance creates a threshold in soil pH at the global scale,  
 891 *Nature*, 540, 567–569, 2016.
- 892
- 893 Smil, V.: *Enriching the Earth: Fritz Haber, Carl Bosch, and the transformation of*  
 894 *world food production*, MIT Press, Cambridge, MA, 2004.
- 895
- 896 Tennant, S.: On different sorts of lime used in agriculture, *Philos. Mag.*, 5, 209–216,  
 897 1799.
- 898
- 899 Tsao, S. S.-E., Surhoff, T. J., Amatulli, G., & Raymond, P. (2025). Agricultural lime  
 900 application across the contiguous United States, 1930–1987 [Data set]. Zenodo.  
 901 <https://doi.org/10.5281/zenodo.15758275>
- 902
- 903 Solly, E. F., Weber, V., Zimmermann, S., Walthert, L., Hagedorn, F., and Schmidt,  
 904 M. W. I.: A critical evaluation of the relationship between the effective cation  
 905 exchange capacity and soil organic carbon content in Swiss forest soils, *Front. For.*  
 906 *Glob. Change*, 3, <https://doi.org/10.3389/ffgc.2020.00098>, 2020.
- 907
- 908 Siqueira, R. G., Moquedace, C. M., Fernandes-Filho, E. I., Schaefer, C. E. G. R.,  
 909 Francelino, M. R., Sacramento, I. F., and Michel, R. F. M.: Modelling and prediction  
 910 of major soil chemical properties with Random Forest: Machine learning as a tool to  
 911 understand soil-environment relationships in Antarctica, *Catena*, 235, 107677, 2024.
- 912
- 913 Ulrich, B.: Natural and anthropogenic components of soil acidification, *Z.*  
 914 *Pflanzenernähr. Bodenkd.*, 149, 702–717, <https://doi.org/10.1002/jpln.19861490607>,  
 915 1986.
- 916
- 917 U.S. Bureau of Mines: *Mineral Resources of the United States*, U.S. Department of  
 918 the Interior, Bureau of Mines, Washington, DC, 1927–1996.
- 919
- 920 U.S. Bureau of the Census: *Census of Agriculture: 1954*, U.S. Govt. Print. Off.,  
 921 Washington, D.C., 1956.
- 922 U.S. Bureau of the Census: *Census of Agriculture: 1959*, U.S. Govt. Print. Off.,  
 923 Washington, D.C., 1961.
- 924 U.S. Bureau of the Census: *Census of Agriculture: 1964*, U.S. Govt. Print. Off.,  
 925 Washington, D.C., 1967.



- 926 U.S. Bureau of the Census: Census of Agriculture: 1969, U.S. Govt. Print. Off.,  
 927 Washington, D.C., 1972.
- 928 U.S. Bureau of the Census: Census of Agriculture: 1974, U.S. Govt. Print. Off.,  
 929 Washington, D.C., 1977.
- 930 U.S. Bureau of the Census: Census of Agriculture: 1978, U.S. Govt. Print. Off.,  
 931 Washington, D.C., 1981.
- 932 U.S. Bureau of the Census: Census of Agriculture: 1982, U.S. Govt. Print. Off.,  
 933 Washington, D.C., 1984.
- 934 U.S. Bureau of the Census: Census of Agriculture: 1987, U.S. Govt. Print. Off.,  
 935 Washington, D.C., 1989.
- 936
- 937 U.S. Geological Survey: Mineral Resources of the United States, U.S. Department of  
 938 the Interior, U.S. Geological Survey, Washington, DC, 1906–1927.
- 939
- 940 U.S. Geological Survey: Minerals Yearbook, vol. 1, U.S. Department of the Interior,  
 941 U.S. Geological Survey, Washington, DC, 1997–2002.
- 942
- 943 Walkinshaw, M., O'Geen, A. T., and Beaudette, D. E.: Soil Properties, California Soil  
 944 Resource Lab, <https://casoilresource.lawr.ucdavis.edu/soil-properties/>, 2023.
- 945
- 946 Weil, R. R., Brady, N. C., and Weil, R. R.: The nature and properties of soils,  
 947 Pearson, Vol. 1104, 2017.
- 948
- 949 West, T. O. and McBride, A. C.: The contribution of agricultural lime to carbon  
 950 dioxide emissions in the United States: Dissolution, transport, and net emissions,  
 951 Agric. Ecosyst. Environ., 108, 145–154, <https://doi.org/10.1016/j.agee.2005.01.002>,  
 952 2005.
- 953
- 954 Wiczorek, M. E.: Area and depth-weighted average of soil pH from STATSGO2 for  
 955 the conterminous United States and District of Columbia [Dataset], U.S. Geological  
 956 Survey, <https://doi.org/10.5066/P95YOSFQ>, 2019.
- 957
- 958 Woodruff, L. G., Cannon, W. F., Eberl, D. D., Smith, D. B., Kilburn, J. E., Horton, J.  
 959 D., Garrett, R. G., and Klassen, R. A.: Continental-scale patterns in soil geochemistry  
 960 and mineralogy: Results from two transects across the United States and Canada,  
 961 Appl. Geochem., 24, 1369–1381, <https://doi.org/10.1016/j.apgeochem.2009.04.009>,  
 962 2009.
- 963
- 964 Xu, D., Zhu, Q., Ros, G., Cai, Z., Wen, S., Xu, M., Zhang, F., and de Vries, W.:



- 965 Calculation of spatially explicit amounts and intervals of agricultural lime  
 966 applications at county-level in China, *Sci. Total Environ.*, 806, 150955,  
 967 <https://doi.org/10.1016/j.scitotenv.2021.150955>, 2022.
- 968  
 969 Zeng, S., Liu, Z., and Groves, C.: Large-scale CO<sub>2</sub> removal by enhanced carbonate  
 970 weathering from changes in land-use practices, *Earth-Sci. Rev.*, 225,  
 971 <https://doi.org/10.1016/j.earscirev.2021.103915>, 2022.
- 972  
 973 Zhang, H.-M., Liang, Z., Li, Y., Chen, Z.-X., Zhang, J.-B., Cai, Z.-C., Elsgaard, L.,  
 974 Cheng, Y., van Groenigen, K. J., and Abalos, D.: Liming modifies greenhouse gas  
 975 fluxes from soils: A meta-analysis of biological drivers, *Agric. Ecosyst. Environ.*,  
 976 340, 108182, <https://doi.org/10.1016/j.agee.2022.108182>, 2022.
- 977  
 978 Zhang, S., Planavsky, N. J., Katchinoff, J., Raymond, P. A., Kanzaki, Y.,  
 979 Reershemius, T., and Reinhard, C. T.: River chemistry constraints on the carbon  
 980 capture potential of surficial enhanced rock weathering, *Limnol. Oceanogr.*, 67,  
 981 <https://doi.org/10.1002/lno.12244>, 2022.
- 982  
 983 Zulauf, C., Schnitkey, G., Paulson, N., and Colussi, J.: Concentration of US crops in  
 984 corn and soybeans: Importance to increasing US production of grains and oilseeds,  
 985 *Farmdoc Daily*, 13, [https://farmdocdaily.illinois.edu/2023/09/concentration-of-us-](https://farmdocdaily.illinois.edu/2023/09/concentration-of-us-crops-in-corn-and-soybeans-importance-to-increasing-us-production-of-grains-and-oilseeds.html)  
 986 [crops-in-corn-and-soybeans-importance-to-increasing-us-production-of-grains-and-](https://farmdocdaily.illinois.edu/2023/09/concentration-of-us-crops-in-corn-and-soybeans-importance-to-increasing-us-production-of-grains-and-oilseeds.html)  
 987 [oilseeds.html](https://farmdocdaily.illinois.edu/2023/09/concentration-of-us-crops-in-corn-and-soybeans-importance-to-increasing-us-production-of-grains-and-oilseeds.html), 2023.
- 988  
 989