

## Interdisciplinary analysis of soil acidification hazard and its legacy effects in Lithuania

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Received: 13 May 2010 – Accepted: 28 June 2010 – Published: 9 July 2010

**Abstract.** An analysis of factors influencing effective soil acidification management is reported. This analysis was conducted simultaneously at both national and local levels. These investigations were accomplished in three stages: (i) validation of acid soil spatial patterns using systems analysis and geoinformation methods; (ii) spatial statistical analysis of soil pH diversity using a statistical grid method; and (iii) development of the concept of soil acidity management. Results indicate the national spatial distribution of topsoil reaction is a natural and stable phenomenon related to Quaternary sub-surface deposits. However, secondary effects of topsoil liming are evident in both spatial and temporal soil reaction patterns.

### 1 Introduction

A major focus of modern global soil science research is the conservation of soil as a fundamental natural resource, in a healthy and high quality condition, capable of yielding economically-viable high quality crops (Karlen et al., 2003). This focus has encouraged the European Commission to recognise soil protection as a policy priority (COM 179, 2002).

Lithuania is located in the humid zone, where mean annual precipitation (748 mm) exceeds mean evapotranspiration (512 mm) (Kilkus et al., 2006) and soil acidification is an ongoing natural process (Bolan et al., 2003). Moreover, acidification is an international soil degradation issue. Thus, research into soil acidification management (SAM) is timely (Eresko, 2005; Ivanov, 2000; Bogdevitch et al., 2005).

SAM is a multifaceted approach utilized by soil science, agro-chemistry, geochemistry and soil geography disciplines. In essence, SAM includes both neutralization of soil acidity and regulation of the acidification of limed soils. The solution depends on a different approach towards the soil as the object of study and the boundaries of anthropogenic effects. A gap has always existed between our knowledge of soil as a natural resource in space and time, and understanding anthropogenic impacts in direct relation to practical solutions.

Experimental investigations of soil acidity management on arable land decreased in most West European and Scandinavian countries several decades ago. Currently, such investigations continue mainly on degraded soils and forest and mountain land-systems (Lundström et al., 2003). In part, this change in focus is because soil is not viewed as a natural amenity, but is used as intermediate media to meet agricultural requirements by the controlled selection and application of agrochemicals to regulate soil acidity. In contrast, in many East and Central European countries, decreases in soil liming and organic fertilizing and associated unbalances in mineral



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fertilizer applications are recurrent problems (Löfgren et al., 1999). With a reduction in agricultural anthropogenic load, agro-ecosystems return to their primeval state through self-regulation. Furthermore, artificially-formed field boundaries are being removed (Rusanov, 2003), causing huge diversity in soil properties even at small scales, where natural soil properties define soil acidity characteristics.

Valuable SAM experience has been gained in Lithuania. In the mid-1960s, acid soils (pH in KCl  $\leq 5.5$ ) covered 41% of agricultural land, which has a national territory of 11 660 km<sup>2</sup>. In some Western and Eastern Lithuanian administrative districts, acid soils covered 70–93% (Savickas, 1973). Following the introduction of large-scale agricultural technologies (1965–1990) the extent of acid soils was successfully decreased to 19% nationally and to 27 and 29% in Western and Eastern Lithuania, respectively. It succeeded due to intensive long-term liming, with applications of dust limestone to 160 000–200 000 hectares per year from 1976. However, as a result of political and economic changes in Lithuania since 1991 and the removal of state support, the extent of liming decreased, falling from 14 400 to 4000 hectares per year between 1993 and 1996. Since 1997, the liming of acid soils has virtually ceased, except on large farms and so soil acidification has accelerated (Mazvila et al., 2004).

To date, the most recent notable achievements in spatial soil acidification research have been in Hungary, Poland, Denmark and Lithuania, where due to periodic soil surveys the databases of national soil reaction data and agrochemical information are collated and soil reaction maps produced (Grybauskas, 1978; Motowicka-Terelak, 1985; Kern, 1987; Madsen and Munk, 1987; Varallyay et al., 1989). These provide a scientific background for developing theories of soil acidification prognosis (Eidukeviciene et al., 2006, 2007; Volungevicius et al., 2006), and the principal mechanism for the interaction of factors determining general spatial regularities of soil pH (Eidukeviciene et al., 2007).

The purpose of this study is to investigate interactions between spatial soil acidity patterns and anthropogenic activities over half a century (~1950–present). This has assisted the development of concepts of soil reaction management employing modern GIS methods.

## 2 Methods

### 2.1 Site, soil characteristics and sampling

Lithuania lies within the area of Pleistocene European Continental glaciation, in the north-west of the East European lowland plain, near the maximum extent of the last glaciation. The topography and associated deposits were formed by the last glaciation (Würm, Weichsel, Vistula, Valdaj). The main soil parent materials are Quaternary deposits, with prevailing glacial till sandy loam, glacio-fluvial sands, glaciolacustrine silty loam and clay. Albeluvisols are the most common

upland soils, while Luvisols prevail on upland slopes (Soil Atlas of Europe, 2005).

A system analysis was developed to investigate natural and anthropogenic factors controlling soil reaction, on an interdisciplinary basis and integrating information on pedology, geology, geography and soil agro-chemistry (Fig. 1). Research was conducted at national and local levels, using a database developed over the last 50 years. At national level, agro-chemical agricultural soil survey data from 1964–2004 were used. These data portray the distribution of acid soils in space and time. At local level, data were abstracted from long-term soil liming field experiments conducted during the period 1949–2006. These data are from Lithuanian Institute of Agriculture research stations in Western and Eastern Lithuania (Vezaiciai and Voke Branches, respectively). The data in Western Lithuania represent moraine deposits (till) Albeluvisols. In Eastern Lithuania, soils are glacio-fluvial Arenosols and Luvisols (Soil Atlas of Europe 2005).

The research has been conducted in three stages:

1. Substantiation of acid soils spatial patterns in Lithuania.
2. Spatial statistical analysis of soil pH diversity.
3. Development of the concept of soil reaction management.

Spatial patterns of acid soils in Lithuania were investigated using system analysis and GIS methods. Performing a spatial multilayer correlation, using GIS, the transformation and co-ordination of geographical information against the LKS-94 co-ordinate system have been achieved using this methodology. Maps of acidic soils (Savickas, 1973), agricultural land pH (Grybauskas, 1978), soil liming priorities (Eidukeviciene, 1993), depth to carbonates (Eidukeviciene and Kudaba, 1976) and soil granulometric composition (Volungevicius et al., 2006) have been compared (Fig. 2).

Spatial statistical analysis of soil pH was carried out on the basis of a modified agricultural land pH map (Eidukeviciene et al., 2006), which shows the distribution of topsoil (0–20 cm depth) pH values for all Lithuania. For statistical analysis of soil pH diversity, a statistical grid method (i.e. 2×2 km, 4 km<sup>2</sup>) was used, generating a database of 16 695 cells. These data were abstracted from agricultural survey data, with one sample per 2–5 ha, depending on landscape variability (Matusevicius, 2005). After Lithuania had been divided into grids, the diversity of soil pH (number of different pH value contours) was evaluated. The soil pH in Lithuania was expressed in diversity points (corresponding to the number of different pH contours). The diversity in Lithuanian soil pH spatial structure is grouped into five index categories: very uniform (index 1–2), uniform (index 3), rather diverse (index 4), diverse (index 5–6) and very diverse (index 7–11). The selection of intervals was related to the binomial distribution structure of the data set. According to the standard deviation of the diversity point from the mean (point 2), the soil pH diversity map was compiled (Fig. 3).

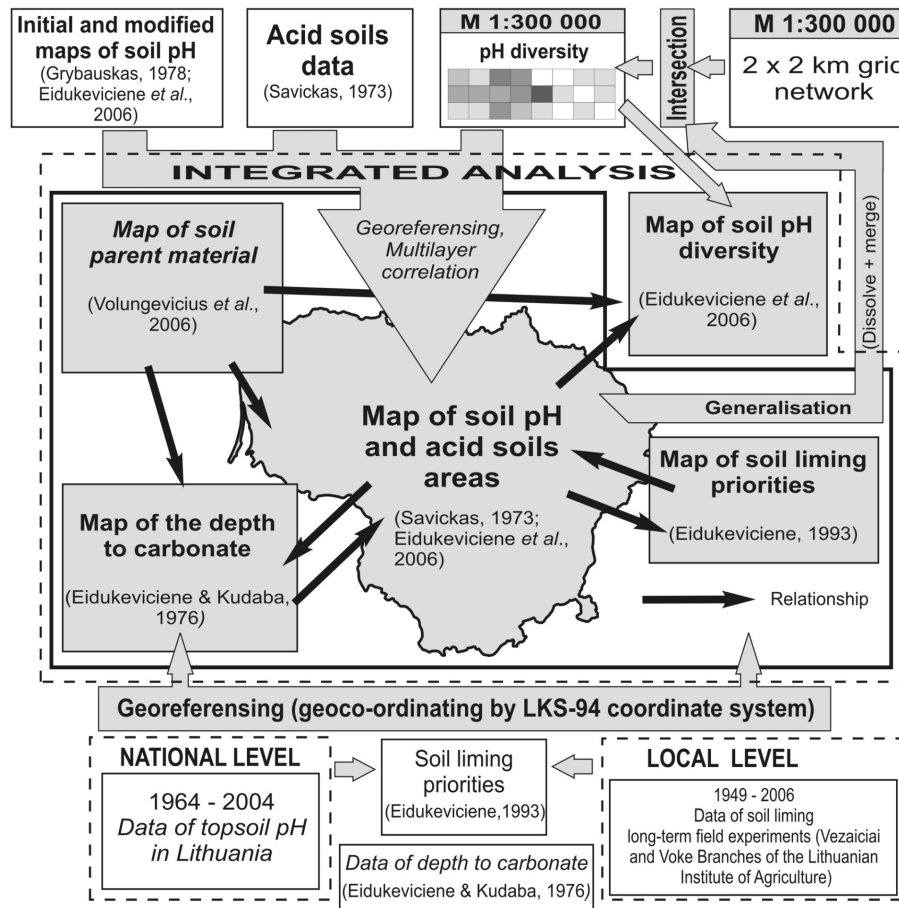


Fig. 1. Methodological scheme of the research.

Changes of the main soil acidity indices (pH and mobile Al) in the entire soil profile under the impact of soil liming have been evaluated from long-term field experiments of soil liming. The liming concept was to apply the amount of lime required to neutralize the soil hydrolytic (potential) acidity (HAC), which was extractable with a neutral unbuffered solution such as KCl. The amount of lime required to neutralize HAC is given by the following equation:

$$\text{Metric tons CaCO}_3 \text{ per hectare} = 0.15 \times \text{HAC} \text{ (meq kg}^{-1}\text{)} \quad (1)$$

Indices of soil pH, using both initial acid soil and various limed soils, were developed based on analysis of 27 individual soil profiles in Voke and Vezaiciai.

## 2.2 Laboratory methods

Soil pH was measured potentiometrically after equilibration with 1.0 N KCl solution. Mobile Al was determined by the Sokolov method (Yagodin et al., 1987). After extraction with 1.0 N KCl solution, a suitable aliquot was titrated with 0.01 N NaOH using phenolphthalein as the indicator for the determination of total exchangeable acidity (H<sup>+</sup>, Al<sup>3+</sup>). In a second aliquot, the acidity from H<sup>+</sup>-ions was determined by the

same titration, after precipitation of the Al<sup>3+</sup>-ions with 3.5% NaF. The quantity of mobile Al was calculated as the difference between the first and second titration. Laboratory data were analysed using Analysis of Variance (ANOVA) (Little and Hills, 1978).

## 3 Results

Conventional liming for over half a century has not abolished national topsoil reaction diversity. Soil reaction (both topsoil and subsoil) at national level and distribution of acid soil areas are fundamentally stable natural phenomena. Outlines of areas of acid soils limed once or twice repeat initial acid soils areas. A trend towards soil acidification is evident in primary acid soil areas, where the extent of liming in the last 15 years has decreased and then stopped. Soil pH diversity in these areas is both low and high. Low soil pH diversity prevails only in specific areas of relatively mature relief, of mainly Middle Pleistocene age (Haplic Albeluvisols, Dystric Planosols and Dystric Luvisols with depth to carbonates > 1.5 m). On areas of initial non-acid soils, which

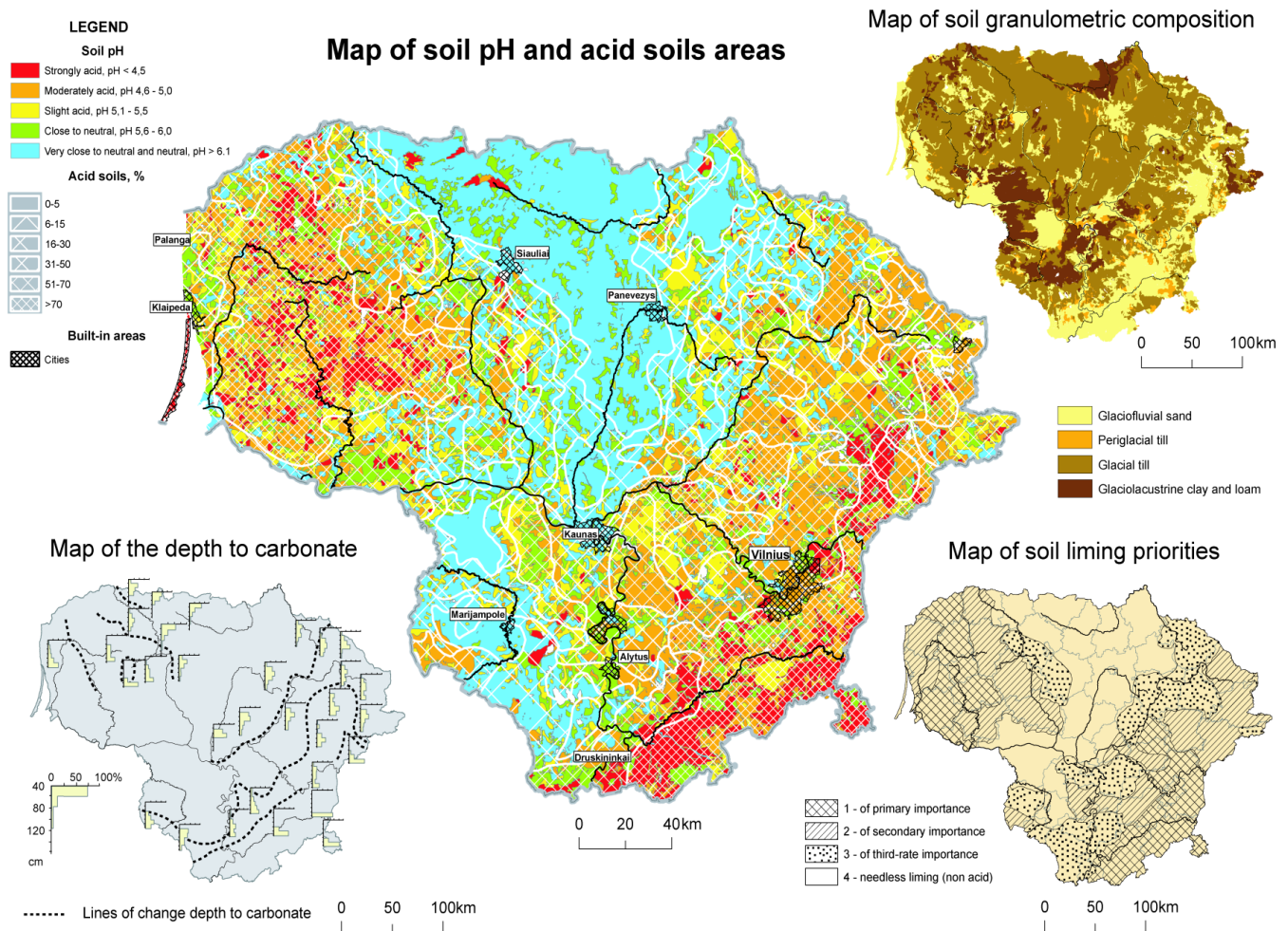


Fig. 2. National map of soil pH.

do not require liming (Gleyic Cambisols, Eutric Cambisols and Calcic Luvisols, depth to carbonates 0.4–0.5 m), soil pH diversity is low. Thus, there are marked spatial variations in liming requirements and priorities need to be identified. Acidification mainly occurs in moraine loam (till) Albeluvisols (Western and Eastern Lithuania), glacio-fluvial sand Arenosols (Eastern Lithuania) and moraine sandy loam Luvisols (South-Eastern Lithuania). Lime applications have ceased on these soils and, consequently, soil acidification is increasing in acid soil territories in Western and Eastern Lithuania, in Albeluvisols, Arenosols and Luvisols, which have a thick acid reaction layer ( $\sim 1.5$  m).

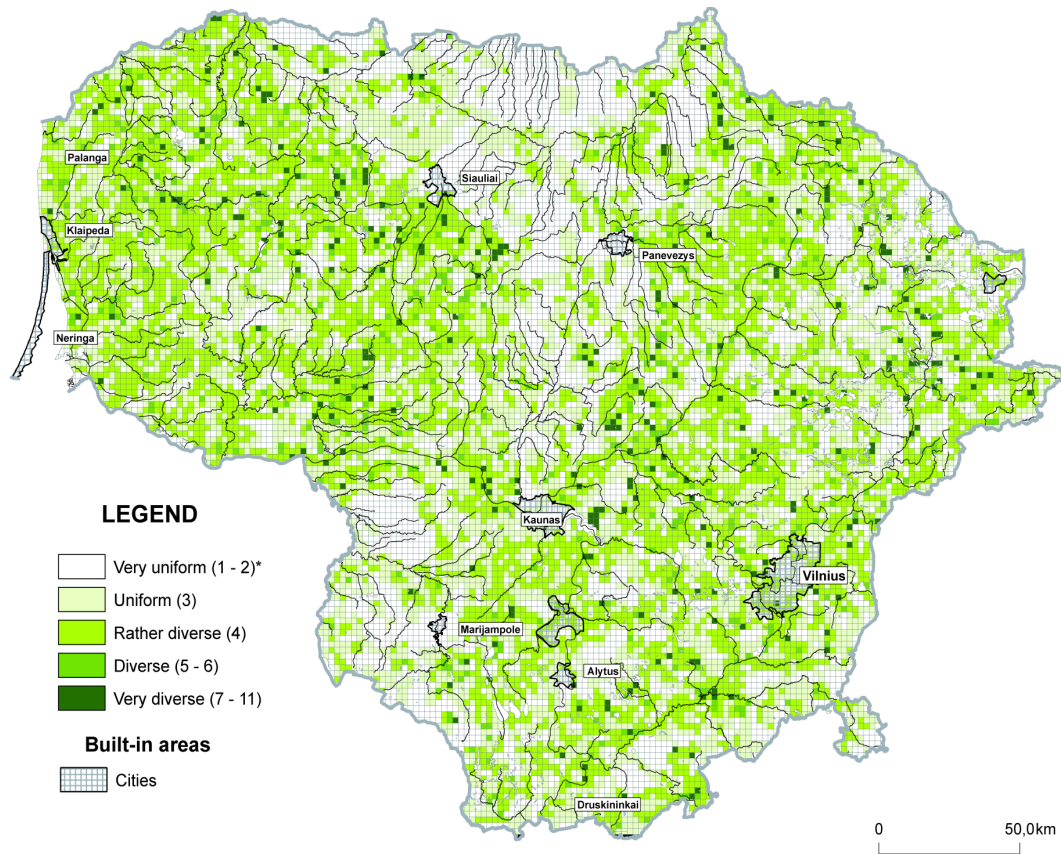
### 3.1 Soil acidity neutralization

Experiments at local level prove that hydrolytic acidity is the main primary liming requirement indicator on moraine sandy loam Albeluvisols and glacio-fluvial sand Luvisols and Arenosols. The most effective means of neutralization is primary liming (single application) at one rate according

to soil hydrolytic acidity. The main acidity indexes in the Albeluvisols topsoil reveal: pH changes from 4.4–6.6 and mobile Al changes from toxic to plants ( $78 \text{ mg kg}^{-1}$ ) to non-toxic ( $4 \text{ mg kg}^{-1}$ ) (Fig. 4). One rate ( $6.6\text{--}7.8 \text{ t ha}^{-1} \text{ CaCO}_3$ ) of lime remains effective (i.e. until soil reaches pH 5.5) in moraine loam for  $\sim 10$  years, while in glacio-fluvial sandy soil one rate of lime ( $6.5 \text{ t ha}^{-1} \text{ CaCO}_3$ ) is effective (until soil reaches pH 5.3) for only 5 years (Fig. 4). Soil pH returns quicker to initial levels in sandy soils than in moraine loams, after 20–22 and 23–28 years, respectively.

Mobile Al was affected by liming more than pH. The amount of mobile Al in soil limed with one rate does not exceed the concentration tolerance of plants ( $20 \text{ mg kg}^{-1}$ ) in glacio-fluvial sandy soil for 12 years and in moraine loam soil for 28 years. Mobile Al in sandy soil increases in the range  $\leq 24 \text{ mg kg}^{-1}$  for 15 years and continues to increase slightly, but even after 34 years, the concentration of mobile Al was half ( $34 \text{ mg kg}^{-1}$ ) of the initial  $66 \text{ mg kg}^{-1}$ . In contrast, in loamy soil, mobile Al increases ( $\leq 30\text{--}35 \text{ mg kg}^{-1}$ ) over 35 years and returns to the initial level in  $\geq 50$  years.





**Fig. 3.** Soil pH diversity in Lithuania.

One rate of lime fertilizers influences acidity indexes in moraine loam even more slowly (35 years) when coarser lime fertilizers, such as tufa (calcareous sediments) or carbonate loam, are used (Ozeraitiene et al., 2006). High fluctuations of mobile Al concentration (from 62 to 110 mg kg<sup>-1</sup>) in moraine loam soil on unlimed treatment possibly resulted from acid precipitation associated with the pollution-intensive industrial period (1960s).

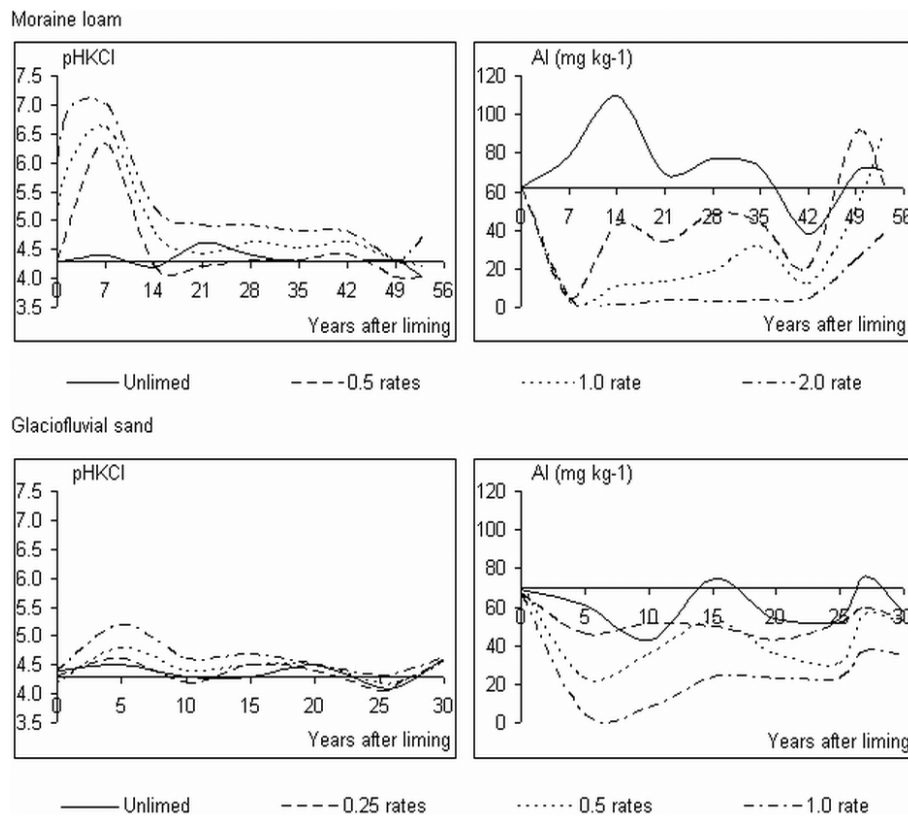
In summary, it is possible to assert that one rate of primary liming according to hydrolytic acidity, essentially influences soil pH up to 10 years and mobile Al for 10–35 years. After primary liming, glacio-fluvial sandy soil acidifies faster than moraine loam.

### 3.2 Management of soil acidity in limed soils

Soil acidification proceeds at different rates: in the initial 7–14 years after liming pH decreases faster (0.1–0.3 pH units per year), but later the process slows down. Acidification and consequent Al release are suppressed for >30 years in sandy and ~50 in loamy soils (Fig. 4). To control soil reaction, repeated and periodic liming cycles and rates on the background of primary liming with one rate should be applied to prevent pH decreased and increased concentrations of toxic Al.

In moraine loamy soil, the recommended pH level of 5.6–6.2 for plants in the topsoil is achieved on the periodic third or fourth liming using a 50-year liming system (primary ×1 rate + repeated ×0.5 rate + periodic ×1 rate every 3–4 years). Soil pH >6.9 can be sustained by repeating periodic liming after application five or six. Periodic liming of glacio-fluvial sandy soil with one rate of dust limestone every 10 years maintains topsoil pH at 5.2–5.4. During periodic liming, mobile Al changes into immobile forms in loams or its concentrations are non-toxic (1.6–2.2 mg kg<sup>-1</sup> in sand) (Fig. 5).

Periodic liming at one rate, followed by different intensities (every 3–4 years and every 10 years) repeated over two decades changes acid soil characteristics both in topsoil and the whole profile to ≤100 cm depth (Eidukeviciene et al., 2001). In moraine loam, maximum changes of acidity indexes (mobile Al from 78 mg kg<sup>-1</sup> to immobile forms and pH from 4.0 to 6.5–6.9) are in topsoil. In EB horizons at 30–50 cm, changes are less (mobile Al from 230 to 80 mg kg<sup>-1</sup> and pH from 3.9 to 5.0–6.2) and changes are insignificant at 50–100 cm depth when soil is limed at ×1 rate every 3–4 years. Liming of glacio-fluvial sand with ×1 rate every 10 years gives maximum effect on the acidity indexes throughout the profile (≤100 cm). Mobile Al changes from 37 to 10 mg kg<sup>-1</sup> and pH from 4.2 to 5.0–5.5. Liming has influenced changes even up to 60 cm. At greater depths, changes



**Fig. 4.** The effect of primary liming on changes in soil acidity indexes.

are related to the effect of carbonate horizons with effervescence having present at 0.8–1.0 m (Eidukeviciene et al., 2001).

Recent findings in forest soils suggest surface horizons are more sensitive to external influences than deeper soils, where soil reaction is more closely related to soil genetic processes (Boruvka et al., 2007). The presented evidence suggests that liming cannot stop or fundamentally change the prevailing soil processes. However, it is possible to maintain pH values at a certain level by using a long-term liming system (primary, repeated and periodic). After primary liming with recommended  $\times 1$  rate, glacio-fluvial sandy soil acidified faster than moraine loam. Despite that, long term systematic liming on glacio-fluvial sand is very effective. Even periodic liming at  $\times 1$  rate every 10 years sustains low acidity reaction and maintains mobile Al below toxic concentrations to  $\leq 100$  cm depth. Yet, in moraine loam soil, three times more intense liming ( $\times 1$  rate every 3–4 years) maintains neutral reaction and immobilizes Al only in the topsoil and decreases its concentration (from 300–150 mg kg<sup>-1</sup>) in EB horizons to 50 cm depth.

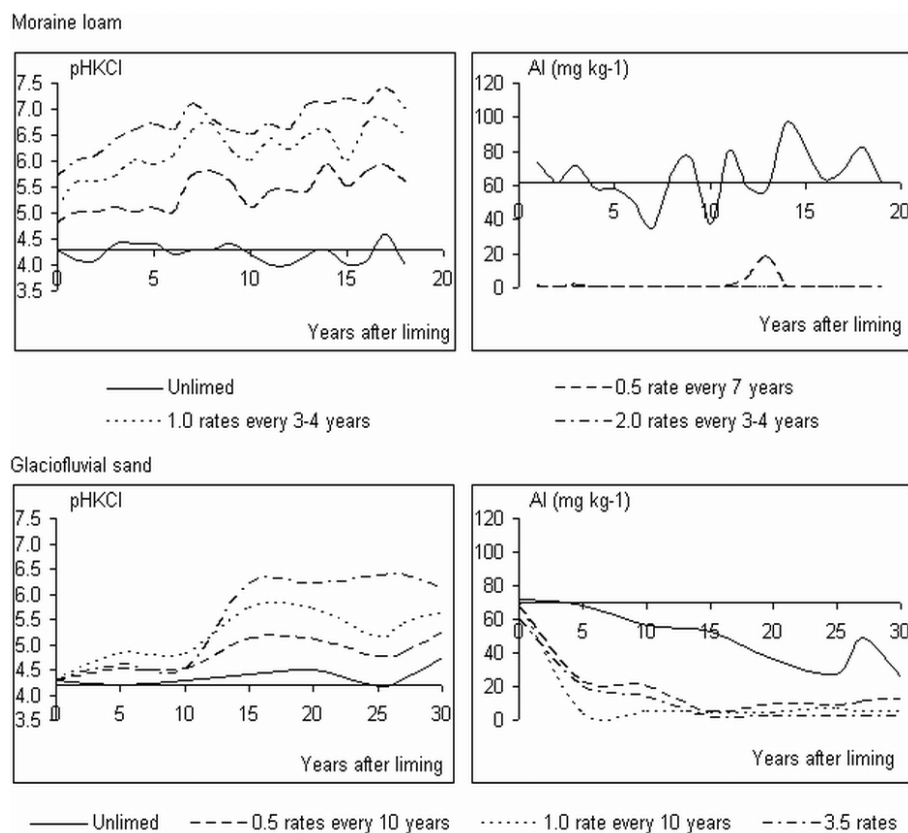
System analysis of 50-years of Lithuanian soil reaction data show that the whole soil system in terms of its thickness and stratigraphy within the topsoil, subsoil and parent materials of Quaternary deposits, plays collective and interacting

roles. Thus, these components should not be considered in isolation. The effect of human economic activity on topsoil in time and space is evident, although the consequences are less important as natural background effects. Both soil processes and the effectiveness of economic activities are subject to natural spatial patterns.

#### 4 Discussion

Awareness of soil acidification hazard issues and timely intervention are the tools that farmers and land managers can use to prevent this land degradation process. Liming at one rate (according to hydrolytic soil acidity) sustains optimum soil pH levels for plant growth. However, we propose additional hypothetical levels: minimal primary liming at  $\times 0.5$  rate and maximum system periodic liming at  $\times 2$  rate every 3–4 years. However, three problems exist: (i) degradation of minimally-limed soil; (ii) degradation of over-limed soil and (iii) substantiation of sustainable maintenance of liming to maintain optimum soil pH.

The problem of cultivation of acid soils four decades ago was based on limited (13-years) experience of long-term field experiments at Vezaiciai, Western Lithuania. Then it was considered that it would be possible to neutralize soil acidity more effectively with two rates of lime application. It was



**Fig. 5.** The effect of periodic liming by different rates of dust limestone on the change of soil acidity indexes.

also hoped that periodic liming would neutralize deeper horizons and decrease acidification rates. However, even then, it was understood that soil cannot be cultivated immediately (Galvydyte, 1968). The 50-year results of long-term liming experiments show these expectations were correct. Although soil researchers could not predict modern problems, these issues did progressively appear. Permanent arable land use has led to the degradation of soil physico-chemical properties. Acidification of permanent arable topsoils is accelerated by the application of fertilizers and intensive leaching of upper soil horizons due to cultivation (Szilassi et al., 2006).

Analysis of Lithuanian soils pH dynamics over half a century under the influence of hypothetical minimal liming level with primary  $\times 0.5$  rate, completed 50 years ago, shows soil acidity neutralization must be an uninterrupted process. Once soil loses its chemical balance it is less resistant to acidification than initially acidic soil. Currently, acidification on soil limed at  $\times 0.5$  rate ( $3.3 \text{ t ha}^{-1} \text{ CaCO}_3$ ) 50 years ago is greater (0.1–0.3 pH units less) than on the initial acid soil (Fig. 4).

Analysis of pH dynamics over half a century under the influence of hypothetical maximum liming level with two rates every 3–4 years according to initial hydrolytic soil acidity is a strategy, which has been applied for some 20 years. Data shows that it results in topsoil pH values of 7.2–7.4 and

pH values of 5.2–5.6 in subsoil to 50 cm depth. However, such notable changes in moraine loam soil pH values cause more intense eluviations of colloidal particles. This results in the decrease of topsoil clay fraction by 2–4% in intensively limed soil compared with unlimed and cause the decrease of soil buffering capacity and faster acidification after liming (Ozeraitiene et al., 2006). From the perspective of soil protection, such intense liming is unjustified and the combinations of intense liming (to maintain soil pH  $\sim 6.7$ ) and excess mineral fertilization ( $\times 3$  NPK) is potentially damaging because it cause the decrease of clay fraction and organic colloidal particles in moraine loam topsoil (Bernotas et al., 2005). Unfavourable consequences of liming include increasing carbon mineralization and nitrate leaching (Hüttle and Schneider, 1998). On the other hand, organic fertilization decreases soil acidification, as the acidifying effect of added ammonium nitrogen is partly compensated by Ca inputs with superphosphate.

Complex soil investigations (agrochemical, physical and microbiological), based on 30-years of liming on moraine loamy soil, show the optimum pH level in moraine loam soils in Western Lithuania is 5.7–6.2, which is achieved by systematic liming with  $\times 0.5$  rates ( $3.8 \text{ t ha}^{-1} \text{ CaCO}_3$ ) every 7 years or  $2.5\text{--}5.5 \text{ t ha}^{-1}$  every 5 years. This necessitates both maintenance liming and the use of the index to determine

lime requirements. Liming concepts are regularly revised and improved in many countries (Tsakelidou, 1995; Matula and Pechova, 2002; Sikora, 2006). In Scandinavia, it is used to determine the rate of lime, not according to pH, but the ratio of cations or nutrient balance data (Wiklander, 1986; Nykänen, 1998).

In Lithuania, a unanimous opinion is not yet agreed. There have been suggestions to use pH, hydrolytic acidity, mobile Al concentration or cation exchange capacity (CEC), depending on the direction of agricultural policy. In Lithuania, there remains a gap between soil liming theory and solving national soil liming problems. In recent years, the influence of socio-economic conditions has increased this knowledge gap.

It has been acknowledged that soil acidification differences in Lithuania, in the absence of liming, are essentially dependent on the genetic diversity of soil acidity in soil profile. Therefore, the concept of differentiated liming is proposed. This prioritizes so-called originally acid soil areas that were present before intensive liming.

## 5 Conclusions

Spatial patterns of topsoil pH in Lithuania are fundamentally controlled by soil parent material. The pH spatial structure of topsoils, limed once or twice, basically echoes initial soil reaction conditions. Topsoil, subsoil and parent material collectively influence soil reaction and should not be treated as separate components. The effect of economic activities on agricultural topsoil in space and time is evident, although the consequences are not as important as natural background effects. Natural and economic processes affect soil in specific areas and the effectiveness of management activities is strongly influenced by pedogenic processes.

In moraine loam, greatest changes of acidity indexes (immobilizing of Al and pH increase from 4.0 to 6.5–6.9) are in topsoil. In EB horizons at 30–50 cm changes are marked less (mobile Al from 230 to 80 mg kg<sup>-1</sup> and pH from 3.9 to 5.0–6.2) and changes are insignificant at 50–100 cm depth when soil is limed at ×1 rate every 3–4 years. Liming of glacio-fluvial sand with ×1 rate every 10 years gives maximum effect on the acidity indexes throughout the profile (≤100 cm). Mobile Al changes from 37 to 10 mg kg<sup>-1</sup> and pH from 4.2 to 5.0–5.5.

*Acknowledgements.* We thank J. Savickas and J. Grybauskas for their deep theoretical foundation of soil liming as a national problem and thorough summary of the first national soil pH investigations in Lithuania. Saulius Marcinkonis gratefully acknowledges the receipt of a Lady Wulfrun Visiting Research Fellowship from the School of Engineering and the Built Environment at the University of Wolverhampton, UK.

Edited by: R. Lasaponara

Reviewed by: I. Bogdevitch and another anonymous referee

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