Global soil acidification impacts on belowground processes

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Abstract

With continuous nitrogen (N) enrichment and sulfur (S) deposition, soil acidification has accelerated and become a global environmental issue. However, a full understanding of the general pattern of ecosystem belowground processes in response to soil acidification due to the impacting factors remains elusive. We conducted a meta-analysis of soil acidification impacts on belowground functions using 304 observations from 49 independent studies, mainly including soil cations, soil nutrient, respiration, root and microbial biomass. Our results show that acid addition significantly reduced soil pH by 0.24 on average, with less pH decrease in forest than non-forest ecosystems. The response ratio of soil pH was positively correlated with site precipitation and temperature, but negatively with initial soil pH. Soil base cations (Ca^{2+}, Mg^{2+}, Na^{+}) decreased while non-base cations (Al^{3+}, Fe^{3+}) increased with soil acidification. Soil respiration, fine root biomass, microbial biomass carbon and nitrogen were significantly reduced by 14.7%, 19.1%, 9.6% and 12.1%, respectively, under acid addition. These indicate that soil carbon processes are sensitive to soil acidification. Overall, our meta-analysis suggests a strong negative impact of soil acidification on belowground functions, with the potential to suppress soil carbon emission. It also arouses our attention to the toxic effects of soil ions on terrestrial ecosystems.

Introduction

Since the mid 20th century, acid rain has become a serious global environmental problem due to rapid industrial development (Blank 1985, Duan *et al* 2016). The main sources of acid and acidifying pollutants are sulfur dioxide (SO_2), nitrogen oxides (NO_x) and ammonia (NH_3) emitted from fossil fuel combustion and agricultural activities (Zhao *et al* 2009, Yang *et al* 2012). Though SO_2 and NO_x emissions have been reduced in Europe and North America, they are increasing in many developing countries due to coal combustion (Gao *et al* 2018). Soil acidification is a natural process, which has been accelerated by increases in N and S deposition associated with human activities (Grieve 2001, Kunhikrishnan *et al* 2016). Human accelerated soil acidification alters biogeochemical cycles and impairs ecosystem function (Stevens *et al* 2010, Liang *et al* 2018). Therefore, understanding the general patterns of ecosystem processes with acid deposition across diverse environments will provide valuable knowledge for predicting future ecosystem dynamics under global change. To date, however, there has been no systematic global synthesis of acid deposition impact on ecosystem functions.

Acid deposition has complex effects on ecosystems, especially for belowground processes. First, more H^+ input to soil along with SO_4^{2-} and NO_3^- induced by acid deposition may directly affect microbial activities (Kuperman and Edwards 1997). Second, H^+ from acid deposition will compete with base cations (e.g. K^+, Mg^{2+}, Ca^{2+}) for replacement, which increase base cation leaching out of soil. This further reduces soil acid buffering capacity (Driscoll *et al* 2003) and nutrient

availability (Likens *et al* 1996). Third, increased soil acidification with continuous acid deposition has the potential to mobilize and release free Al^{3+} and Fe^{3+} to soil solution. This accumulation of toxic elements in topsoil may eventually impair root growth and microbial activity (Godbold *et al* 1988, Kochian 1995, Poschenrieder *et al* 2008), which will consequently reduces soil respiration.

The rate and form of acid deposition, soil type, environmental factor, and ecosystem type all may regulate the effects of acid deposition on soil processes. Normally, acid deposition rate should be a major factor to drive soil acidification (Vanhala *et al* 1996). Different acid forms, e.g. H_2SO_4 and HNO_3, may also contribute to the variable impacts of acid deposition due to their different adsorption mechanism. NO_3^- is adsorbed only through electrostatic attraction, while SO_4^{2-} can be specially adsorbed through ligand exchange, especially in variable charge soils (Curtin and Syers 1990, Guadalix and Pardo 1991). This special adsorption may lead to a release of hydroxyl ions, which could neutralize a part of the acids and retard soil acidification to some extent. Furthermore, soil type is a significant contributor to regulating soil acidification response. It is expected that soils with different initial pH may go through different acidification buffering phases (Bowman *et al* 2008). Soil with a lower pH generally experiences greater acid-weathering, which makes less sensitive to external acid input (De Vries *et al* 1989, Zhu 2017). High precipitation accelerates the leaching of soil cations and further aggravates acidification (Lapenis *et al* 2004, Ling *et al* 2007). Low temperature possibly depresses litter decomposition (Oulehle *et al* 2011, Liang *et al* 2013), leading to litter accumulation and then weakening soil acidification magnitude (Aerts 1997). The influence of these abiotic and biotic factors in combination finally causes different ecosystem response to acid deposition. It is a challenge but essential to quantify the influences of those factors on the soil acidification impacts across different experiments with different application rates and acid agents to soils with varying buffering capacities.

Here, we compiled a global dataset (304 observations) from 49 case studies and performed a meta-analysis to quantify belowground process dynamics in response to experimental acid addition. Specifically, we addressed the following questions: (1) How have various belowground processes respond to acid addition at global scale? (2) What are the main controlling factors for the responses of belowground processes?

Materials and methods

Data compilation

Web of Science, Google Scholar and China National Knowledge Infrastructure were searched for peer-reviewed publications on experiments dealing with acid deposition and ecosystem function. The searched keywords were: (acid deposition or S deposition or simulated acid rain) AND (soil cations (e.g. Na^+, Mg^{2+}, Ca^{2+}), soil nutrient (e.g. SOC, STN), soil respiration, fine root biomass, microbial biomass). The following criteria were employed to screen appropriate studies for analysis: (1) only acid addition experiments in the field were included, with the treatment duration lasting at least one growing season; (2) The control and acid addition treatments had to experience the same climate and soil condition; (3) examined variables were required to be clearly described by their means, sample sizes and standard deviation.

To acquire as many observations as possible, we gathered the data at each peak biomass stage of the growing season during all measurement years. If several studies with different vegetation types or environmental conditions (i.e. annual temperature or precipitation) were reported in an article, each study was considered to be independent. Table-form data were directly extracted, while graph-form data were obtained by the Engauge Digitizer software (Free Software Foundation, Inc., Boston, MA, USA). If climate variables could not be obtained from the papers, we used the latitude and longitude of each study to extract these data from a global database (http://worldclim.org/). Soil type data were acquired from the FAO database (http://fao.org/). Finally, a global dataset was established with 49 independent studies from 45 papers (figure S1 is available online at stacks.iop.org/ERL/14/074003/mmedia). This dataset covered the area with latitude range from 23.15 to 69.75° N and elevation range from 10 to 1200 m. Mean annual temperature varied from −2 °C to 21.4 °C, and precipitation from 130 to 2400 mm. Ecosystems included forest, grassland and peatland, but we sorted them into two groups for analysis (forest and non-forest). This is due to the lack of data from grassland and peatland ecosystems. Experimental duration spanned 1 to 14 years.

In our dataset, most data were related to belowground processes. Response variables included soil cations (K^+, Na^+, Mg^{2+}, Ca^{2+}, Zn^{2+}, Mn^{2+}, Al^{3+}, Fe^{3+}), soil nutrient (SOC-soil organic C, DOC-dissolved organic C, STN-soil total N, soil NH_4, soil NO_3, soil available P, soil C:N), soil respiration, fine root biomass, and microbial biomass (MBC-microbial biomass C, MBN-microbial biomass N, Bac-bacterial biomass and Fun-fungal biomass). Furthermore, our dataset also involved other background data, such as longitude, latitude, elevation, climate factors (i.e. temperature and precipitation).

Meta-analysis

As described in previous studies (Hedges *et al* 1999, Lu *et al* 2011), we employed meta-analysis techniques to evaluate the impacts of acid addition on ecosystem

belowground processes. Effect sizes of acid treatment were calculated as equation (1):

$$\log(RR) = \log(\bar{X}_{\text{treatment}}/\bar{X}_{\text{control}}) = \log\bar{X}_{\text{treatment}} - \log\bar{X}_{\text{control}}, \quad (1)$$

where $\bar{X}_{\text{treatment}}$ and $\bar{X}_{\text{control}}$ are the mean values in acid addition and control treatments, respectively.

Effect sizes and their subsequent inferences in meta-analysis may be influenced by how individual observations are weighted (Mueller *et al* 2012, Ma and Chen 2016). According to previous studies (Wu *et al* 2011, Ma and Chen 2016) and our analysis, the weighted method based on variance assigns extreme importance to individual effect sizes, with a result that overall effect is mostly determined by a small number of extreme observations. Thus, we calculated a weighting factor (w) based on the sample size in each experiment as follows (Adams *et al* 1997, Pittelkow *et al* 2015, Zhang *et al* 2018).

$$w = \frac{n_{\text{control}} \times n_{\text{treatment}}}{n_{\text{control}} + n_{\text{treatment}}}, \quad (2)$$

where n_{control} and $n_{\text{treatment}}$ are the sample size of variables in control and acid treatment, respectively.

Linear mixed effect model fitted with the Restricted Maximal Likelihood was utilized to analyze the impacts of acid addition on belowground processes as follows ('lme4' R package) (Bates *et al* 2015):

$$\ln(RR) = \beta_0 + \pi_{\text{study}} + \varepsilon, \quad (3)$$

where β_0, π_{study} and ε are coefficient, the random effect of 'study' and sampling error respectively. The possible autocorrelation among observations within each study was explicitly accounted by the random effect of study (Chen and Chen 2018). Linear, power and quadratic functions were applied to examine the relationships between log(RR) of belowground processes (i.e. soil cations-K^+, Na^+, Mg^{2+}, Ca^{2+}, Zn^{2+}, Mn^{2+}, Al^{3+}, Fe^{3+}; soil nutrient-SOC, DOC, STN, NO_3^-, NH_4^+, AP, soil C:N; soil respiration; fine root biomass; microbial biomass) and log(RR) of soil acidification (soil pH versus acid addition level), MAT or MAP. We selected the best bivariate relationships based on the Akaike Information Criterion (AIC). If the difference in AIC among multiple models was less than two, the simple model was selected. If the difference in AIC was larger than two, the model with lower AIC was chosen (Wagenmakers 2003). Furthermore, we found that the residuals of most models did not follow a normal distribution with the Shapiro–Wilk's test. Thus, we applied a nonparametric bootstrap analysis to estimate the effect sizes and their 95% confidence interval (CI) using the 'boot' package (Davison and Hinkley 1997, Canty and Ripley 2012). In addition, we analyzed the relationships of belowground processes with both acid addition level and soil pH change, and found that the results were consistent between two analyses. To include as much data in analysis as possible, we mainly presented the results with the soil pH analysis in our main text, but put those with the acid level analysis in the supplementary material.

All independent variables were scaled to ensure the comparability between the results. For ease of interpretation, we converted the results of log (RR) as a percentage of belowground process responses to acid treatment (i.e.100 × ($10^{\log(RR)} - 1$)). All statistical analyses and figure plotting were performed in R version 3.4.3 (R Core Team 2017).

Results

Effects of acid treatment on belowground processes

At global scale, acid addition significantly reduced soil pH by 0.24 on average (figures 1, 5). Similarly, acid addition caused a significant decrease in soil base cations, such as Na^+, Mg^{2+} and Ca^{2+}. In contrast, acid treatment increased soil Al^{3+} and Fe^{3+} by 22.7% and 48.6%, respectively (figures 1, 5). Moreover, acid addition further reduced soil carbon processes significantly, with a lower decrease in MBC (9.6%) than fine root biomass (19.1%) and soil respiration (14.7%). For microbial community, bacterial biomass showed a significant decrease (16.4%), whereas fungal biomass was not sensitive (figures 1, 5). Acid addition mostly had no impact on soil nutrient, such as SOC, DOC, STN, NH_4–N, NO_3–N, available P or soil C: N.

Across the acid addition gradient, soil pH was significantly reduced when acid addition rate was higher than 5 kmol H^+ ha^{-1} yr^{-1} (figure 2). Below this level, soil pH did not show any significant response. Furthermore, we found similar relationships of belowground processes with acid addition level versus soil pH change. To be concise, here we mainly presented those results with the pH analysis. For soil cations, soil Na^+, Mg^{2+} and Ca^{2+} reduced linearly with decreasing soil pH (figure 3). By contrast, soil Al^{3+} and Fe^{3+} showed a positive relationship with soil acidification (figure 3). With respect to soil nutrient, soil acidification significantly suppressed soil NO_3–N and soil C: N, but promoted soil NH_4–N and available P (figure 3). Soil respiration and microbial biomass carbon displayed a positive relationship with soil acidification (figure 3). Moreover, soil acidification linearly reduced bacterial biomass, whereas enhanced fungal biomass (figure 3).

For different forms of acid addition, our results demonstrated that most belowground processes showed similar responses to both addition of $H_2SO_4 + HNO_3$ and H_2SO_4 alone. Moreover, no significant difference of soil NO_3^- occurred between these two acid forms. There was a positive response of soil NH_4^+ caused by H_2SO_4 addition, but no response under $H_2SO_4 + HNO_3$ addition (figure S3).

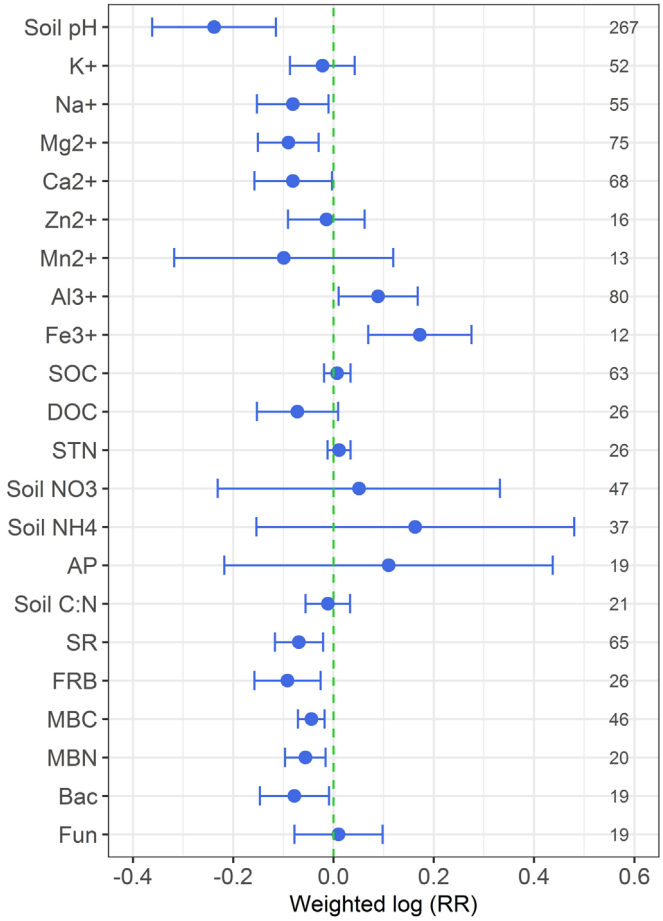

Figure 1. Effects of acid addition on soil pH, soil cations (K^+, Na^+, Mg^{2+}, Ca^{2+}, Zn^{2+}, Mn^{2+}, Al^{3+}, Fe^{3+}), soil nutrient (SOC-soil organic C, DOC-dissolved organic C, STN-soil total N, Soil NO_3–NO_3–N, Soil NH_4–NH_4–N, AP-available P, soil C:N), soil respiration (SR), fine root biomass (FRB) and microbes (MBC-microbial biomass C, MBN-microbial biomass N, Fun-fungal biomass, Bac-bacterial biomass). Error bars represent 95% confidence intervals (CIs). The dotted line was drawn at weighted mean log (RR) = 0. The simple size for each variable was shown on the right. The effect of acid addition is considered significant if the 95% CI does not cover zero. Based on the formula of $\log_{10}(H^+_{treatment}/H^+_{control}) = \log_{10}(H^+_{treatment}) - \log_{10}(H^+_{control})$, soil pH change by acid addition was represented as ($pH_{control} - pH_{treatment}$). Notably, the data in the graph were presented by $pH_{treatment} - pH_{control}$.

Ecosystem type affecting acid addition effects

Between different ecosystems, forests had a lower reduction in soil pH (0.15) than non-forest ecosystems (0.96). In forests, acid addition posed a significant impact on most belowground processes, negatively affecting soil Mg^{2+}, Ca^{2+}, soil respiration, fine root biomass, MBC, MBN and bacterial biomass but positively influencing soil Fe^{3+}, NH_4–N, available P and fungal biomass (figure S4a). In non-forest ecosystems, acid addition only decreased soil Na^+ by 29.5% and increased soil Al^{3+} by 87.1% (figure S4b). In addition, among different soil types, acrisols, kastanozem, luvisol and podzol showed a significant pH reduction, whereas anthrosol, calcisol and cambisol had no significant response (figure 2).

Environmental factors regulating acid addition effects

The response ratio of soil pH showed a negative relationship with initial soil pH (figure 4(a)), but a positive relationship with site temperature and precipitation (figures 4(b), (c)). High temperature tended to lessen the response magnitudes of soil Al^{3+}, NO_3–N, NH_4–N and available P to acid addition, but to enhance that of fine root biomass (figure S5b). More precipitation intensified the effect of acid treatment on soil Mg^{2+} and fine root biomass, while it attenuated the impact on soil Al^{3+}, NH_4–N and fungal biomass (figure S5a).

Discussion

Soil itself is a buffer system for external H^+ input. When H^+ input exceeds the maximum of soil buffer capacity, it will cause soil acidification. By synthesizing the results from global acid addition experiments, we found that acid addition significantly reduced soil pH by 0.24 unit (figures 1, 5). A similar reduction in pH occurred, with a decrease of 0.26 pH unit, reported in a global synthesis of nitrogen addition experiments (Tian and Niu 2015), but it is lower than a decrease of 0.63 unit reported from Chinese northern grasslands over the last two decades (Yang *et al* 2012). Soil acidification became significant when the H^+ addition

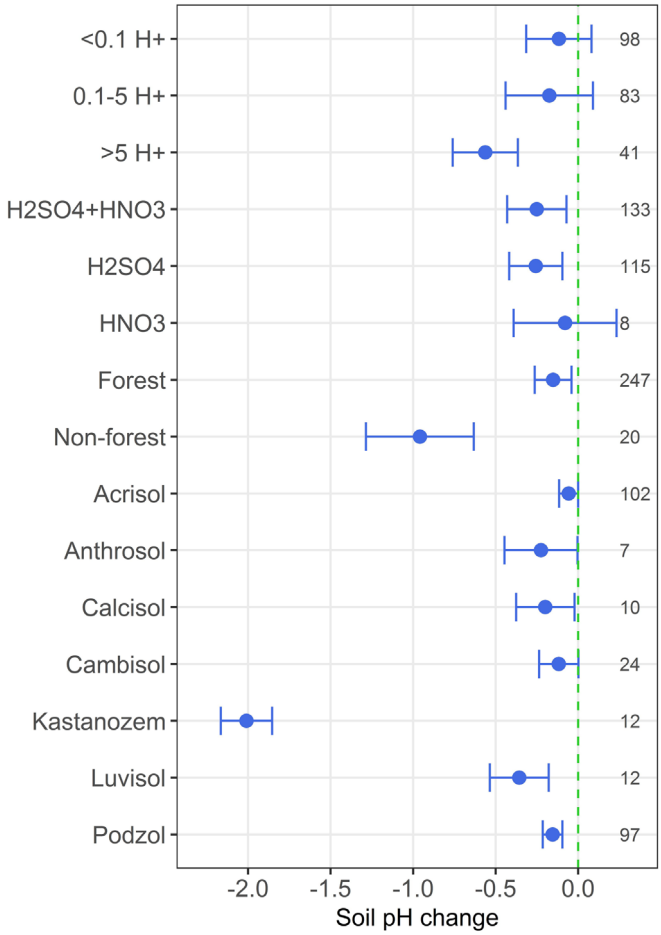

Figure 2. Soil pH change under acid addition experiments with different acid levels, acid forms, ecosystem types or soil types. The dotted line was drawn at soil pH change = 0. The sample size for each variable was shown on the right. Error bars represent 95% confidence intervals (CIs). The effect of acid addition is considered significant at $\alpha = 0.05$ if the 95% CI does not cover zero. The unit of H^+ (acid addition rate) is kmol ha^{-1} yr^{-1}.

rate was more than 5 kmol ha^{-1} yr^{-1}, which indicates a threshold level of acid deposition driving acidification (Liao and Jiang (2002)). The similar responses of belowground processes between different forms of $H_2SO_4 + HNO_3$ and H_2SO_4 addition imply a dominant effect of soil acidification and little impact by N-fertilizer. However, different soil types showed different sensitivities to acid addition (De Vries *et al* 1989). The largest soil pH decrease in Kastanozem is likely because its carbonate is susceptible to H^+ input (Bowman *et al* 2008, IUSS Working Group WRB 2015). This may lead to the larger decrease of soil pH in non-forests than forest ecosystems.

Acid addition-induced soil acidification further altered soil nutrient availability. The increase of soil available P with soil pH reduction indicates that much H^+ input by acid addition mainly promotes the release of phosphate from Fe or Al binding compound (Barrow and Shaw 1979, Barrow 2017). In addition, we found that soil NH_4^+ increased with a decrease in soil pH, likely due to the inhibition of nitrification and plant N uptake (Kemmitt *et al* 2005, Vanguelova *et al* 2007, Chen *et al* 2013). Furthermore, more H^+ inhibition of nitrification can suppress the transformation of soil NH_4^+ to NO_3^-, resulting in a negative relationship of soil NO_3^- with acid addition level (figure S2) (Chen *et al* 2013).

In line with our expectation, soil acidification caused a significant reduction in fine root biomass, which is mainly due to the following reasons. First, acid deposition accelerated the leaching of soil base cations (Mg^{2+}, Ca^{2+}), further reducing their availability (Vanhala *et al* 1996, Pennanen *et al* 1998, Chen *et al* 2013, Chen *et al* 2015). This may directly reduce plant uptake for these essential elements, resulting in plant nutrient deficiency and then limiting primary productivity (Kochian 1995, Van Den Berg *et al* 2005, Vanguelova *et al* 2007, Chen *et al* 2013). Second, once base cations have been depleted, soil may release and accumulate toxic ions like Al^{3+} (Bowman *et al* 2008). In this study, we indeed found that acid addition led to a significant increase in Al^{3+} and Fe^{3+}. Increasing Al^{3+} can reduce root nutrient absorption, and much absorption of Al^{3+} will interfere with plant physiological processes and finally cause Al^{3+} toxicity (Ulrich *et al* 1980). These together could explain the fact that root biomass was significantly reduced by acid deposition (Hahn and Marschner 1998, Li *et al* 2018).

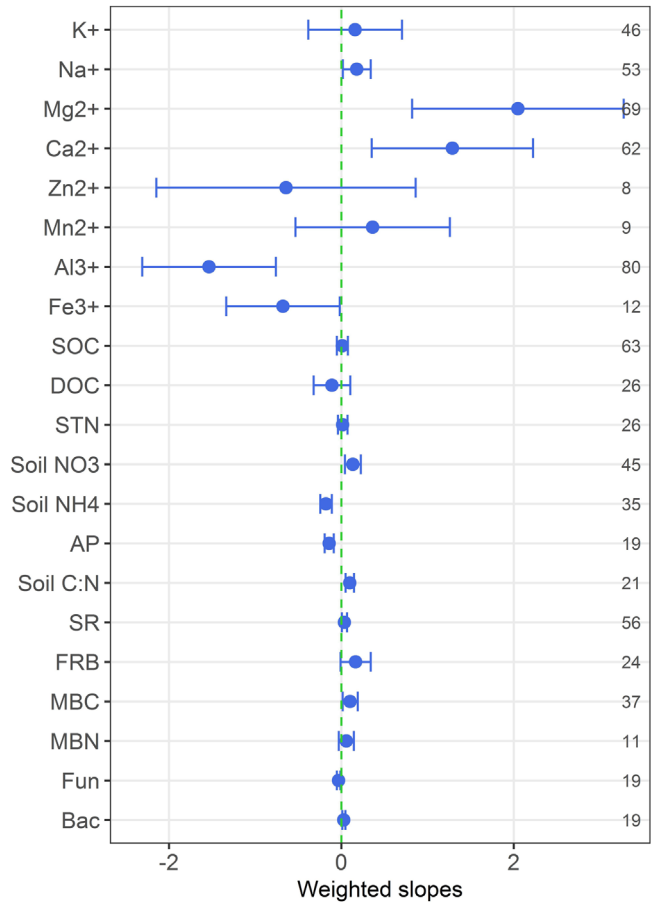

Figure 3. Linear slopes in the relationship between the response ratio of soil pH and soil cations (K^+, Na^+, Mg^{2+}, Ca^{2+}, Zn^{2+}, Mn^{2+}, Al^{3+}, Fe^{3+}), soil nutrient (SOC-soil organic C, DOC-dissolved organic C, STN-soil total N, Soil NO_3–NO_3–N, Soil NH_4–NH_4–N, AP-available P, soil C:N), soil respiration, fine root biomass or microbes (MBC-microbial biomass C, MBN-microbial biomass N, Fun-fungal biomass, Bac-bacterial biomass). If the 95% CI does not overlap the slope = 0, it means a significant response in soil belowground processes caused by acidification.

Acid deposition also induced a significant decrease in bacterial biomass, but had no effect on fungal biomass (figures 1, 5). As shown in the summary figure (figure 5), acid addition-induced soil acidification and the ion toxicity (Al^{3+} and Fe^{3+}) were two key factors to affect soil microbes (Chen *et al* 2013). The decline in bacteria biomass and no change in fungal biomass may be attributed to the fact that fungi are less sensitive to acidification and more tolerant to H^+ and Al^{3+} when compared to bacteria (Rousk *et al* 2009, Aliasgharzad *et al* 2010, Strickland and Rousk 2010). For instance, due to the difference in cell wall structure between fungi and bacteria (Myrold and Nason 1992), fungi can store excess H^+ in the vacuoles and then extrude it into the environment (Kuperman and Edwards 1997). For the Al^{3+} toxicity under acid deposition, on the one hand, its high concentration may directly decrease microbial biomass due to its toxicity to microbial cells (Pina and Cervantes 1996). On the other hand, higher level of Al^{3+} perhaps reduces plant substrate inputs to soil (e.g. dead root, root exudate), further decreasing the conversion efficiency of plant C into microbial biomass C (Pietri and Brookes 2008, Oulehle *et al* 2018).

Soil respiration, which consists of autotrophic respiration (R_a) and heterotrophic respiration (R_h), is one of the largest carbon effluxes in terrestrial ecosystems (Kuzyakov 2006, Luo and Zhou 2006). The C release through R_a or R_h is associated with belowground C pools, such as root biomass, soil organic C (SOC) and microbial biomass C (MBC) (Zhou *et al* 2014). Several mechanisms for root and microbial processes may help explain the decline in soil respiration under soil acidification (figure 5). First, more H^+ likely reduces microbial physiology and biomass, and then depresses soil heterotrophic respiration (Riggs and Hobbie 2016). Second, the nutrient limitation of microbial growth due to soil base cation loss (e.g. Mg^{2+}, Ca^{2+}) (Bowman *et al* 2008, Oulehle *et al* 2018) and other ion (e.g. H^+, Al^{3+}) toxicity (Tian and Niu 2015) tend to reduce soil heterotrophic respiration. Moreover, we also found a negative effect of acid addition on fine root biomass, which should cause a decrease in autotrophic respiration and thus soil respiration (Davidson *et al* 2006, Liang *et al* 2013).

Acid deposition impacts on belowground processes varied with environmental factors. Soil pH decreased

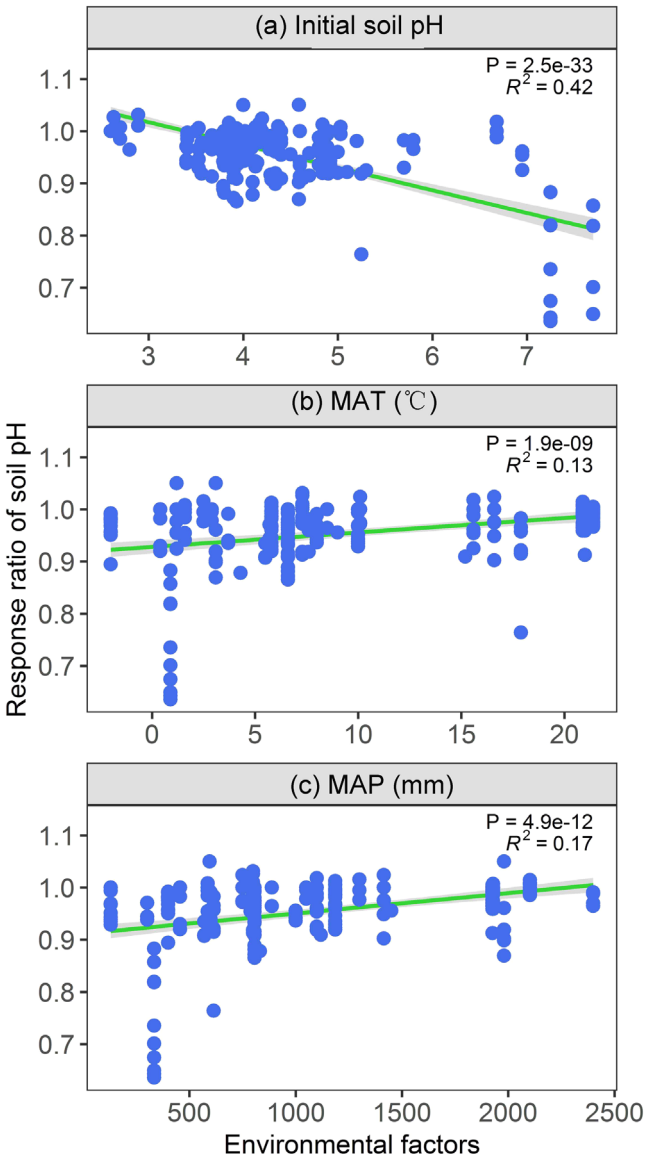

Figure 4. Relationships in the response ratio (*RR*) of soil pH with initial soil pH (a), mean annual temperature (MAT), (b) or mean annual precipitation (MAP), (c).

more at sites with high initial pH (figure 4(a)), which is likely because soils have different acid buffering capacities depending on the initial pH. It is generally accepted that the exchange capacity of soil cation is: trivalent cations (Fe^{3+} > Al^{3+}) > divalent cations (Mn^{2+} > Zn^{2+} > Ca^{2+} > Mg^{2+}) > monovalent cations (K^+ > Na^+). High valence cations will be replaced to buffer soil acidification only when low valence cations are mostly depleted. Based on the charge equivalent principle, soil with high initial pH is more vulnerable to be acidified. Contrary to our expectation, soil pH reduced less in sites with higher precipitation (figure 4(c)). Although more precipitation is expected to promote soil acidification by accelerating cation leaching, sites with high precipitation had a low initial soil pH in our study, which were less vulnerable to be acidified (figure 4(c)). In a word, our findings emphasize the interactive effects of acid deposition and environmental factors on soil acidification.

Overall, this study has important implications for soil biogeochemical cycles. We found that acid addition significantly reduced the exchangeable base cations of Na^+, Mg^{2+}, Ca^{2+} in soils. Similar to the result of N-induced soil acidification (Tian and Niu 2015), acid deposition induced soil acidification process goes through different buffering stages. The decline in soil pH is accompanied by a depletion of soil base cations. Once base cations have been depleted, soil reaches toxic levels because of Al^{3+} release. A significant increase in soil free Al^{3+} has been already detected under acid deposition, suggesting that soil in terrestrial ecosystem has begun to enter Al^{3+} buffering stage. All these alert our attention to the danger of the coming soil acidification and its buffering stages of toxic Al^{3+} and Fe^{3+}.

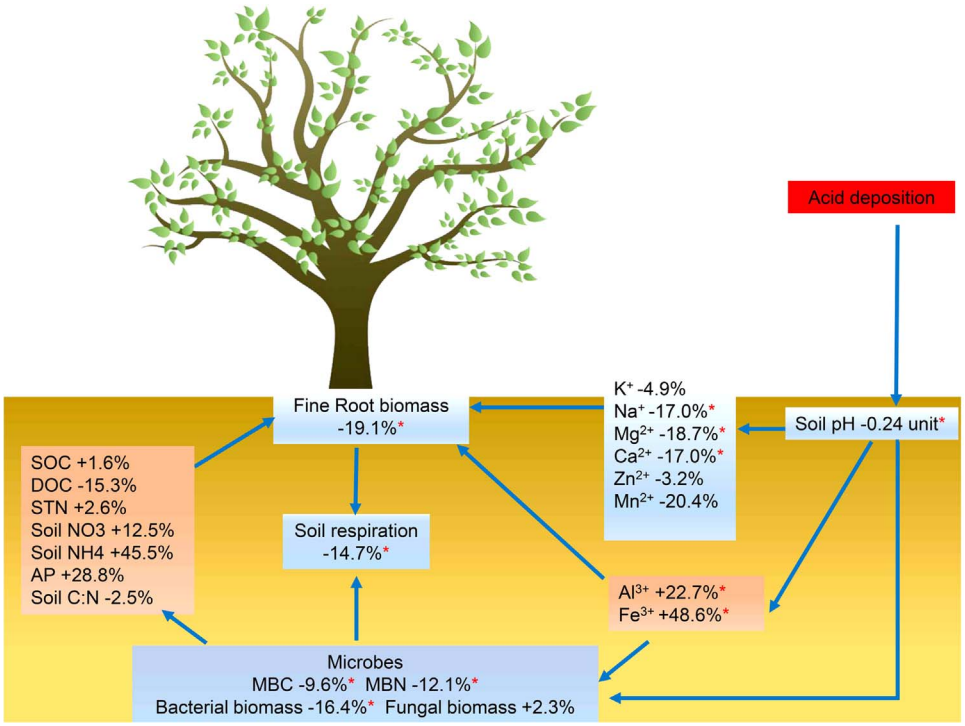

Figure 5. Conceptual figure summarizing the relative changes in belowground processes under acid addition.

Conclusion

Our synthesis revealed that belowground processes were sensitive to acid deposition at global scale and across different ecosystems. Global soil acidification is currently in a transition stage from base cation (Ca^{2+}, Mg^{2+}, K^{+}) to non-base cation buffering (Al^{3+}, Fe^{3+}). This calls our attention to the toxic effects of soil ions on terrestrial ecosystems. Moreover, acid deposition further caused a decline in microbial biomass, fine root biomass and soil respiration, suggesting that the inhibition of soil carbon emission will substantially change soil carbon balance and its feedback to climate change. However, it is difficult to predict the magnitude of soil acidification with acid deposition and its impacts on belowground processes, mainly due to the complex relationships of acid effects with diverse environmental factors (soil properties and climate). Overall, this meta-analysis provides the first global viewpoint on linking belowground processes with soil acidification under acid deposition.

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