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Proton production from nitrogen transformation drives stream export of base cations in acid-sensitive forested watersheds

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ABSTRACT

The biogeochemical cycles of nitrogen (N) and base cations (BCs), (i.e., K^+, Na^+, Ca^{2+}, and Mg^{2+}), play critical roles in plant nutrition and ecosystem function. Empirical correlations between large experimental N fertilizer additions to forest ecosystems and increased BCs loss in stream water are well demonstrated, but the mechanisms driving this coupling remain poorly understood. We hypothesized that protons generated through N transformation (PPR_N)—quantified as the balance of NH_4^+ (H^+ source) and NO_3^- (H^+ sink) in precipitation versus the stream output will impact BCs loss in acid-sensitive ecosystems. To test this hypothesis, we monitored precipitation input and stream export of inorganic N and BCs for three years in an acid-sensitive forested watershed in a granite area of subtropical China. We found the precipitation input of inorganic N (17.71 kg N ha^{-1} $year^{-1}$ with 54% as NH_4^+–N) was considerably higher than stream exported inorganic N (5.99 kg N ha^{-1} $year^{-1}$ with 83% as NO_3^-–N), making the watershed a net N sink. The stream export of BCs (151, 1518, 851, and 252 mol ha^{-1} $year^{-1}$ for K^+, Na^+, Ca^{2+}, and Mg^{2+}, respectively) was positively correlated (r = 0.80, 0.90, 0.84, and 0.84 for K^+, Na^+, Ca^{2+}, and Mg^{2+} on a monthly scale, respectively, $P < 0.001$, $n = 36$) with PPR_N (389 mol ha^{-1} $year^{-1}$) over the three years, suggesting that PPR_N drives loss of BCs in the acid-sensitive ecosystem. A global meta-analysis of 15 watershed studies from non-calcareous ecosystems further supports this hypothesis by showing a similarly strong correlation between $\sum$BCs output and PPR_N ($r = 0.89$, $P < 0.001$, $n = 15$), in spite of the pronounced differences in environmental settings. Collectively, our results suggest that N transformations rather than anions (NO_3^- and/or SO_4^{2-}) leaching specifically, are an important mediator of BCs loss in acid-senstive ecosystems. Our study provides the first definitive evidence that the chronic N deposition and subsequent transformation within the watershed drive stream export of BCs through proton production in acid-sensitive ecosystems, irrespective of their current relatively high N retention. Our findings suggest the N-transformation-based proton production can be used as an indicator of watershed outflow quality in the acid-sensitive ecosystems.

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1. Introduction

Nitrogen (N) and the dominant base cations (BCs), (i.e., K^+, Na^+, Ca^{2+}, and Mg^{2+}), include essential macronutrients for plants and critical elements for ecosystem functions. The importance of N limitation to primary production and ecosystem functions has been demonstrated in a wide variety of terrestrial and aquatic ecosystems (Vitousek and Howarth, 1991; Reich et al., 2006; LeBauer and Treseder, 2008). However, excess N inputs, due to elevated N deposition or experimental N addition, have led to N saturation in many temperate forests (Aber et al., 1998; Lovett and Goodale, 2011). The consequences of N saturation include an increase in nitrate leaching (Kahl et al., 1993) and a decrease in the abundance of dominant forest understory and groundcover species (Talhelm et al., 2013). Ca^{2+}, Mg^{2+} and K^+ also play a vital role in many physiological processes, such as

Abbreviations: BCs, base cations; PPR_N, proton production rate from N transformation.

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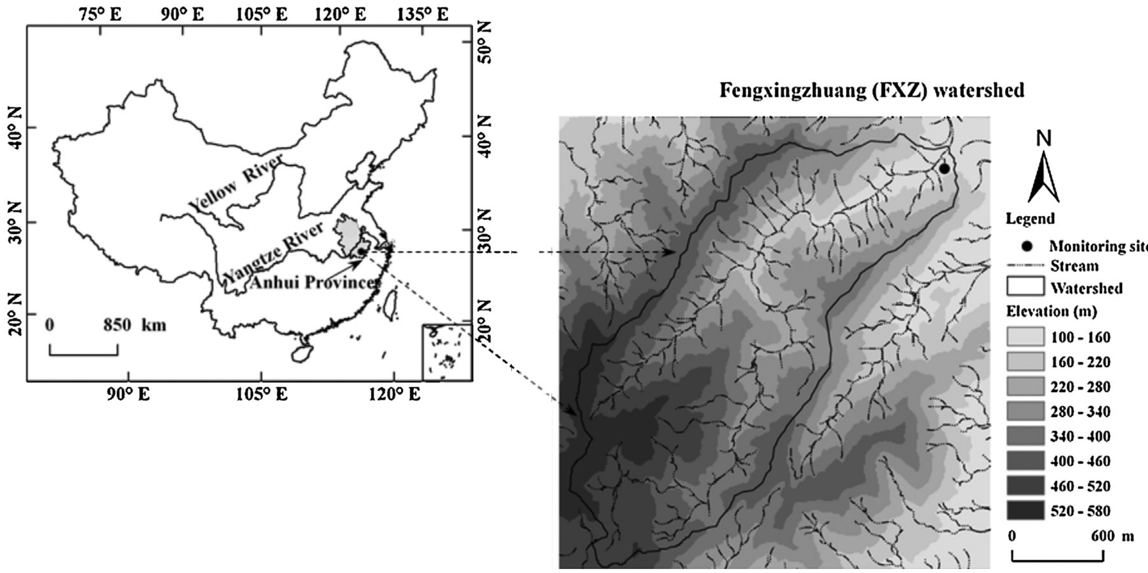

Fig. 1. Location and digital elevation model of the Fengxingzhuang forested watershed. The left side is the standard map of People's Republic of China with South China Sea Islands in the right bottom square and the right side is the digital elevation model of the studied Fengxingzhuang forested watershed.

photosynthesis, energy storage, stomatal pore closure, and cell signaling (McLaughlin and Wimmer, 1999). Na^{+} does not have a specific metabolic role in plants, but it along with other BCs, participate in exchange reactions that buffer against changes in soil and water acidity (Likens et al., 1998; Houle et al., 2006). Decreases in BCs pools and availability in soils could ultimately result in decreased productivity, reduced tolerance to environmental stresses (e.g., drought, freezing, and pathogen attacks), and a decline in the ecosystem's ability to buffer changes in acidity.

Previous studies have shown that large experimental N additions can significantly accelerate BCs leaching from terrestrial ecosystems (Van Miegroet and Cole, 1984; Chung and Zasoski, 1993; Horswill et al., 2008; Mitchell and Smethurst, 2008) and cause a reduction in available BCs in less than five years (Lucas et al., 2011). Large punctual N fertilizer additions are expected to result in rapid buildup of soil N and consequently lead to N saturation (Kahl et al., 1993), which would support intensive nitrification and accelerate nitrate and BCs leaching (Aber et al., 1998). However, extrapolating these results to natural ecosystems is difficult because chronic atmospheric N deposition occurs at a much lower rate than the large punctual fertilizer additions in the experiments above. It remains unclear whether the chronic atmospheric N deposition influences BCs status in natural ecosystems.

N deposition is likely to increase along with the general trend of increase in reactive N creation (Galloway et al., 2008), and this will have important impacts on ecosystem properties or processes. It is well known that N deposition could cause soil acidification through internal proton production by N transformation, especially in acid-sensitive ecosystems (Van Breemen et al., 1983, 1984; De Vries and Breeuwsma, 1987; Gundersen and Rasmussen, 1990). But the effects of proton production on ecosystem BCs status have not been evaluated. We hypothesize that protons generated through N transformation (PPR_N) will impact BCs loss in stream water in the acid-sensitive forested watersheds. To test this, we monitored precipitation input and stream export of inorganic N and BCs for three years in an acid-sensitive forested watershed in a granite area of subtropical China. In addition, we performed a global meta-analysis of related watershed studies from non-calcareous ecosystems that had simultaneous BC and N input and output data. Our objectives were to: (1) establish the relationship between stream export of BCs and PPR_N; (2) suggest the mechanisms that control the response of BCs loss in stream water to PPR_N; and (3) evaluate the indication value of N-transformation-based proton production in the acid sensitive ecosystems.

2. Materials and methods

2.1. Study area

The Fengxingzhuang is a 3.59 km^2 forested watershed with elevations between 100 and 575 m and an outlet at 30°33′ N, 118°2′ E in Jinxian county, southern Anhui Province, in south China (Fig. 1). It is a representative of a typical acid-sensitive ecosystem because of its non-calcareous parent material and the low acid-neutralizing capacity of its soils (Huang et al., 2011, 2012a,b, 2013 Yang et al., 2013). This region has a typical Asian monsoon climate characterized by mild springs with frequent, light rainfall, hot and humid summers with strong southeasterly monsoon sea breezes, and relatively cool and dry autumns and winters influenced by the northwestern monsoonal winds. The mean annual air temperature and mean annual precipitation are 16.5 °C and 1585 mm, respectively. The coniferous forest covers nearly 80% of the Fengxingzhuang watershed with an understory of shrubs, vines, grasses and ferns. Soils are derived from granitic rocks that formed in the Late Yenshan movement of the Late Cretaceous Period (Gu et al., 2003) and contain quartz and plagioclase (with a minor mica fraction) as primary minerals and kaolinite, vermiculite and chlorite as secondary minerals in clay-size fraction (Yang et al., 2013). The soils have high sand content (>50%), low cation exchange capacity (<15 cmol kg^{-1}) and low pH (<5.5), throughout the shallow soil profile (less than 50 cm), and are classified as Lithic Dystrudepts (Inceptisols) and Lithic Udorthents (Entisols) according to soil taxonomy (Soil Survey Staff, 1999).

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