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Increasing importance of nitrate-nitrogen and organic nitrogen concentrations in bulk and throughfall precipitation across urban forests in southern China

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ABSTRACT

Atmospheric nitrogen (N) deposition is an increasingly serious threat to forest ecosystems requiring urgent global actions. While it is well-known that developing countries are experiencing rapid urbanization, an understanding of its effects on N composition and sources in atmospheric deposition is only beginning to emerge. We measured dissolved inorganic N (DIN), dissolved organic N (DON) and total dissolved N (TDN) concentrations in bulk precipitation (BP) and throughfall precipitation (TP) across four seasons in forest ecosystems along the more urbanized river (Bailongjiang; BJR) and the less urbanized river (Wulongjiang; WJR) in Fuzhou coastal city. Concentrations of all N forms were greatly enhanced in BJR forests than in WJR forests, suggesting increased anthropogenic N pollution in forests along the BJR that, in turn, can cause serious N cycle perturbations. While precipitation N concentrations over the BJR forests were primarily influenced by pollutants from fossil fuel combustion activities (ammonium-nitrogen; NH_4^+-N/nitrate-nitrogen; NO_3^--N = 0.77), those over the WJR forests were collectively influenced by pollutants from fossil fuel combustion and agricultural activities (NH_4^+-N/NO_3^--N = 1.14). The DON contributed 22–38% of N to the TDN across seasons and rivers and was positively related to other N forms, indicating that DON is steadily becoming a vital component in atmospheric N deposition especially in anthropogenically N-polluted atmospheres. Summer was characterized by precipitation with lower N concentrations, highlighting increased dilution effect and decreased anthropogenic N emissions. Nitrogen concentrations were more elevated in TP than in BP, suggesting that tree canopies augment atmospheric N deposition to forest soils. Generally, soil and foliar TN and $\delta^{15}N$ were positively related to precipitation TDN, indicating that increased atmospheric N deposition enhances forest ecosystem N availability. Our results demonstrate that urban areas particularly in developing countries are not only NH_x but also NO_x emission and deposition hotspots and emphasize the necessity of including them in future global N budgets and N pollution abatement initiatives.

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1. Introduction

Concern about global nitrogen (N) biogeochemical cycle perturbations has been growing significantly in recent decades. The rapid increase in urbanization-induced N emissions due to increases in anthropogenic activities within urban ecosystems including N fertilizer application during lawn management and fossil fuel combustion in industrial and transportation sectors has contributed much to these perturbations (Baker et al., 2001; Zhao et al., 2009; Huang et al., 2015). These increases in urbanization-induced N emissions along with other anthropogenic-induced N emissions primarily in agricultural systems have more than tripled the global rates of atmospheric reactive N deposition from 31.6 to 103 Tg N per year between 1860 and early 1990s (Galloway et al., 2004). They also forecasted that these current rates will increase to approximately 195 Tg N per year in 2050.

Although there are few positive effects of N deposition particularly on plant growth, food production and carbon sequestration that have been documented in some ecosystems (Thomas et al., 2010; Ti et al., 2012; Fleischer et al., 2013; Griepentrog et al., 2014), there are many negative effects of elevated N deposition on ecosystems and humans that are increasingly recognised e.g. acidification of soils, eutrophication and loss of biodiversity (Richter et al., 2005; Diaz and Rosenberg, 2008; Huang et al., 2012; Clark et al., 2013; Stevens et al., 2018). Despite the fact that urban areas around the world are thought to emit considerable amounts of N that might be caused in large part by higher concentration of anthropogenic activities in these areas, there have been little atmospheric N deposition studies within and near urban areas. While some of these studies have indicated a decrease in N deposition along an urban-suburban-rural gradient (Kuang et al., 2011; Huang et al., 2012; Rao et al., 2014), others have shown that suburban areas are experiencing elevated N deposition than urban and rural counterparts (e.g. Fang et al., 2011). Therefore, the effect of urbanization on N deposition represents a knowledge gap and that an accurate understanding of the spatio-temporal patterns of atmospheric N inputs in ecosystems along gradients of urbanization is becoming increasingly important for evaluating its effects and for suggesting its control measures.

Inorganic and organic N species are the two components of N deposited from the atmosphere into urban ecosystems. Because of the significant contribution of the former component (ammonium; NH_4^+-N plus nitrate; NO_3^--N) on the total N deposition, most previous urban studies have focused on its deposition and its biogeochemical cycling (e.g. Huang et al., 2012; Rao et al., 2014; Decina et al., 2017). However, the few recent studies that have investigated organic N deposition across urban ecosystems worldwide have alerted on its increasing contribution to total atmospheric N deposition (de Souza et al., 2015; Araujo et al., 2015; Izquieta-Rojano et al., 2016; Decina et al., 2018). Thus, it is necessary to measure this component in N deposition studies to clearly understand its deposition rates, abundance and sources as well as its importance on total atmospheric N input budgets.

In addition to atmospheric organic N deposition measurements, it is necessary to quantify the potential effects of the tree canopy on organic and inorganic N inputs and budgets to urban ecosystems, in particular forests. Several studies have shown that tree canopy-exchange processes can transform the deposited NH_4^+-N and NO_3^--N through various mechanisms including foliar uptake, leaching and the action of microbes, epiphytic bryophytes and lichens (Kopáček et al., 2009; Ponette-González et al., 2010; Adriaenssens et al., 2011; Fenn et al., 2013; Templer et al., 2015). Although urban studies have indicated that the rates of throughfall (inputs underneath tree canopies) are higher than those of bulk (inputs underneath open sky) deposition (e.g. Juknys et al., 2007; Fang et al., 2011; Decina et al., 2018), throughfall measurements are often neglected in many urban N deposition studies (Li et al., 2012; de Souza et al., 2015; Araujo et al., 2015). As trees are increasingly being planted in urban areas to enhance sustainable provision of ecosystem services to urbanites including erosion control, nutrient cycling and climate regulation, it is important to investigate the potential effects of their canopies on atmospheric N inputs in urban ecosystems.

Urbanization-induced anthropogenic activities i.e. N fertilizer application and fossil fuel combustion have been increasing rapidly in China over the past few decades (Huang et al., 2015). These activities have increased N emissions, placing China among the largest N emitter globally (Liu et al., 2011, 2013). Despite the elevated emissions of N in urban areas across the country, there have been few studies on the rates and trends of atmospheric N deposition to urban ecosystems. Like in other countries, organic and throughfall N inputs have been largely neglected in many of these studies (Fang et al., 2011; Li et al., 2012; Huang et al., 2012, 2015), irrespective of their importance to total atmospheric N deposition. Some of these studies have also shown that urbanization in China is rapidly changing the composition and main sources of atmospheric inorganic N deposition, from agricultural-related NH_4^+-N dominance to fossil fuel combustion-related NO_3^--N dominance (Zhao et al., 2009; Wang et al., 2013; Huang et al., 2013, 2015). However, it is still unknown whether this change is universal across China's urban areas.

This study reports the measurements of atmospheric N deposition in forest stands along the two urbanized coastal rivers in a subtropical Fuzhou city in southern China between 2017 and 2018. Forest ecosystems along these rivers are likely to be receiving elevated rates of atmospheric N inputs due to increasing urbanization-induced anthropogenic activities in recent years. The following were the specific objectives of this study: (1) to investigate different N forms i.e. ammonium-nitrogen (NH_4^+-N), nitrate-nitrogen (NO_3^--N), NH_4^+-N/NO_3^--N ratio, dissolved inorganic nitrogen (DIN), dissolved organic nitrogen (DON) and total dissolved nitrogen (TDN) in bulk and throughfall precipitation (hereafter BP and TP, respectively) across forest ecosystems of the more urbanized (Bailongjiang; BJR) and the less urbanized (Wulongjiang; WJR) rivers in the context of their concentrations, spatio-temporal distributions and potential sources and (2) to correlate precipitation TDN concentrations with soil and foliar TN and $\delta^{15}N$ data taken from our previous study (Mgelwa et al., 2019a). The correlated data were

collected during the summer of 2017 from similar forest stands. The findings of the current study will broaden our knowledge on the patterns of different N forms in precipitation, including those of understudied organic N, and the associated effects on N biogeochemical cycle across urban forests.

2. Materials and methods

2.1. Study sites

This study was conducted in southern China's Fuzhou coastal city (25°15′–26°39′ N & 118°08′–120°31′ E) (Xue et al., 2012). The total city area is 11,968 km^2 (Xue et al., 2012) with a mean elevation of 84 m (Cai et al., 2015) and a population in excess of 7 million residents (Xue et al., 2012). Fuzhou experiences a typical sub-tropical monsoon climate with abundant rainfall. Its mean annual rainfall is 1346 mm (Zheng et al., 2006) of which most (>70%) falls in the wet season between spring and summer. The mean annual temperature of the city is 19.6 °C (Zheng et al., 2006). Six forest stands were selected along the two Minjiang River Estuary branches i.e. 31.99 km Bailongjiang (BJR) and 34.8 km Wulongjiang (WJR) rivers. Among these forest stands, three i.e. Bailongjiang River Upstream (BJRU), Bailongjiang River Midstream (BJRM) and Bailongjiang River Downstream (BJRD) belonged to the BJR and three i.e. Wulongjiang River Upstream (WJRU), Wulongjiang River Midstream (WJRM) and Wulongjiang River Downstream (WJRD) belonged to the WJR. We also used these forest stands in our recent studies (Mgelwa et al., 2019a,b). Additionally, the Bailongjiang river is more urbanized compared with the Wulongjiang river (Liu et al., 2016).

2.2. Sample collections and laboratory analysis

BP and TP samples were collected at three BJR forest stands and three WJR forest stands from 2017 in July (summer) and November (autumn) to 2018 in February (winter) and May (spring). Only one sampling event was conducted per season. Each sample was collected using a 25-cm diameter polyethylene funnel, attached to a 10-L polyethylene bottle. Sample collectors were mounted in a supporting stand ~1 m above the ground level to avoid contamination of the samples from splashed soil and water. The funnels were covered with a 3 mm mesh to keep out vegetal debris, insects and bird-droppings. Collectors for BP (three per site) were located underneath open sky and those for TF (three per site) were placed underneath tree canopies. After rain event, the bottles were thoroughly shaken before the samples were transferred to 1-L plastic bottles. The samples were immediately sent to the Forest Ecology and Stable Isotope Research Center at Fujian Agriculture and Forestry University in China, where they were filtered (Whatman #42 filter paper) and stored at −20 °C until analysis. After each sampling event, the collectors were washed properly with tap water, rinsed thoroughly using distilled water and then dried at room temperature. Moreover, the precipitation and temperature data for June 2017–May 2018 period were obtained from Fuzhou meteorological station (Fig. 1). The concentrations of ammonium-nitrogen (NH_4^+-N) and nitrate-nitrogen (NO_3^--N) in BP and TP samples were analysed with a discrete wet chemistry analyzer (Westco SmartChem® 200, Analyzer Medical System S.r.l., Italy) and their sum (NH_4^+-N + NO_3^--N) were considered as the total dissolved inorganic nitrogen (DIN) concentrations. Total dissolved nitrogen (TDN) concentrations were determined using a Shimadzu TOC/TN analyzer (TOC-L CPH, Shimadzu, Japan). The concentrations of dissolved organic nitrogen (DON) were calculated as the difference between TDN and DIN i.e. [DON = TDN − DIN = TDN − (NH_4^+-N + NO_3^--N)].

Soil and foliar field sampling and laboratory analysis procedures have already been explained in detail in Mgelwa et al. (2019a) and are therefore only briefly explained here. Both soil and foliar samples were collected in the summer of 2017 from six forest stands located at the upstream, midstream and downstream areas of the Bailongjiang and Wulongjiang rivers.

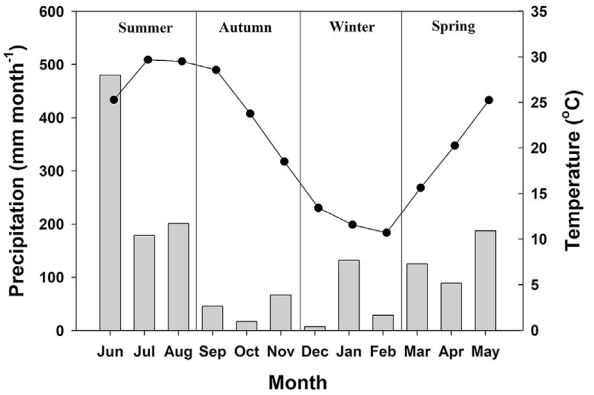

Fig. 1. Monthly total precipitation (gray bar) and mean temperature (dotted line) from June (2017) to May (2018) over two rivers in Fuzhou coastal city.

As mentioned earlier, precipitation samples used in the current study were also collected from these forest stands. For foliar sampling, composite samples were formed in each forest stand by pooling sufficient number of fully developed and healthy foliage collected randomly across five different individuals of each dominant species. About three to five dominant species, each representing a single composite sample, were randomly chosen per forest stand. For soil sampling, six replicates were collected across two (0–10 cm and 10–20 cm) soil depths in each forest stand. A total of eight soil cores were pooled to form each replicate. Each composite sample was stored in a labelled and sealed bag immediately after collection and all samples were then taken to the laboratory within 12 h of collection. In the laboratory, while foliar samples were washed and oven-dried at 65 °C for 48 h, soil samples were hand sieved through a 2-mm mesh sieve and air-dried at room temperature. Oven-dried foliar and air-dried soil samples were ground in a ball mill (JXFSTPRP-64, Jingxin Co., Ltd, China). For the determinations of soil and foliar N concentrations and $\delta^{15}N$ values, finely ground soil (30 mg) and foliar (4 mg) samples were therefore analysed on an isotope ratio mass spectrometer (IRMS) (IsoPrime 100, Isoprime Ltd., UK), linked to a CN elemental analyzer (vario MICRO cube, Elementar, Germany).

2.3. Data analysis

All data were analysed in R 3.3.1 Statistical Software (R Development Core Team, 2017) whereas all plotting were performed in SigmaPlot Version 14.0 (Systat Software, Inc., San Jose California USA). Prior to analyses, the data were examined for violations of normality and homogeneity of variance assumptions using the Shapiro–Wilk and Levene tests, respectively. Differences in the concentrations of BP and TP nitrogen variables (i.e. NH_4^+-N, NO_3^--N, NH_4^+-N/NO_3^--N, DIN, DON and TDN) both among rivers (i.e. BJR and WJR) as well as between seasons (i.e. summer, autumn, winter and spring) were compared with a one-way ANOVA, which was followed by a post-hoc Tukey's HSD test when the differences in means were significant. Two-way ANOVA was applied to assess the effects of river × season interactions on BP and TP nitrogen variables. Potential relationships between precipitation, soil and foliar N variables were determined using Pearson's correlation analysis. All data are presented as mean (standard deviation) unless otherwise stated. The level of significance for all statistical tests was set at $p < 0.05$.

3. Results

3.1. Patterns of nitrogen concentrations in BP and TP across rivers and seasons

Both river and season had a significant effect on the concentrations of BP and TP nitrogen forms; however, the river × season interactions affected primarily the concentrations of BP nitrogen forms (Tables 1 and 2). The concentrations of NH_4^+-N and NO_3^--N were significantly higher in the BJR forests than in the WJR forests both in BP (NH_4^+-N, $p = 0.01$; NO_3^--N, $p = 0.003$) and TP (NH_4^+-N, $p = 0.002$; NO_3^--N, $p < 0.001$). Of the two inorganic N forms, NO_3^--N contributed more N to TDN in the BJR forests but not in the WJR forests, where the contributions of NH_4^+-N and NO_3^--N were almost similar in both BP and TP (Fig. 2).

The NH_4^+-N + NO_3^--N increased whereas the NH_4^+-N/NO_3^--N decreased considerably from the WJR forests to the BJR forests (all $p < 0.001$); specifically, the sum and the ratio of NH_4^+-N and NO_3^--N concentrations were on average 1.79 mg L^{-1} and 1.14 in the WJR forests and 3.56 mg L^{-1} and 0.77 in the BJR forests, respectively (Tables 1 and 2). BJR forests have 3 times more DON in BP ($p < 0.001$) and 2 times more DON in TP ($p < 0.001$) than WJR forests. The DON accounted for higher percentages of the TDN across the two rivers, in particular BJR, where the contributions were up to 29% in BP (Fig. 2). Moreover, the TDN increased by 41% in BP and 53% in TP from the WJR towards the BJR (both $p < 0.001$; Tables 1 and 2).

The summer was characterized by significantly lower NH_4^+-N concentrations than the winter ($p = 0.02$) in BP and autumn ($p = 0.002$), winter ($p = 0.01$) and spring ($p < 0.001$) in TP. Concentrations of NO_3^--N in summer were the lowest across seasons, considerably lower compared with the spring ($p = 0.004$) in BP and the three other seasons in TP (all $p < 0.05$). Generally, NO_3^--N contributed more N to TDN than NH_4^+-N especially in TP over the course of the seasons (Fig. 2). The DIN concentrations revealed seasonal patterns similar to those of the NO_3^--N concentrations (Tables 1 and 2). There were no remarkable differences in the ratio of NH_4^+-N/NO_3^--N across seasons (both $p > 0.05$) except between spring and winter in BP, with much higher ratio being observed during the winter than the spring ($p = 0.01$).

The concentrations of DON in summer were substantially lower, on average, than those in autumn ($p = 0.01$) and spring ($p = 0.002$). However, the contributions of DON to the TDN were more prominent during the summer, where the contributions of DIN were somewhat lower (Fig. 2). Also the summer has 3 times less TDN compared with the spring in BP ($p = 0.009$) and 2 times less TDN than the autumn, winter and spring in TP (all $p < 0.05$). Furthermore, while no considerable difference in the ratio of NH_4^+-N/NO_3^--N was observed between BP and TP ($p = 0.82$), the concentrations of NH_4^+-N, NO_3^--N, DIN, DON and TDN were significantly higher in the TP compared with the BP (all $p < 0.001$; Tables 1 and 2). The NH_4^+-N, NO_3^--N, DIN and DON concentrations accounted for 32%, 41%, 73% and 27% of the TDN in BP and 31%, 44%, 75% and 25% in TP, respectively. Additionally, the NO_3^--N, DIN, DON and TDN in BP and DON in TP were substantially affected by the river × season interactions (all $p < 0.05$; Tables 1 and 2).

Table 1
Concentrations of ammonium-nitrogen (NH_4^+-N), nitrate-nitrogen (NO_3^--N), NH_4^+-N/NO_3^--N ratio, dissolved inorganic nitrogen (DIN), dissolved organic nitrogen (DON) and total dissolved nitrogen (TDN) in bulk precipitation across seasons in forest stands of the Bailongjiang (BJR) and Wulongjiang (WJR) rivers.

River	[a]Stand	Season	NH_4^+-N (mg L^{-1})	NO_3^--N (mg L^{-1})	Ratio	DIN (mg L^{-1})	DON (mg L^{-1})	TDN (mg L^{-1})
BJR	BJRU	Summer	0.60 (0.03)	0.71 (0.01)	0.85 (0.03)	1.31 (0.04)	0.69 (0.04)	2.00 (0.08)
		Autumn	1.41 (0.04)	1.84 (0.16)	0.77 (0.04)	3.25 (0.20)	1.37 (0.08)	4.62 (0.28)
		Winter	1.46 (0.94)	1.37 (0.18)	1.02 (0.53)	2.83 (1.11)	1.10 (0.31)	3.93 (1.40)
		Spring	1.10 (0.33)	2.52 (0.52)	0.43 (0.05)	3.61 (0.84)	2.29 (0.49)	5.90 (1.32)
	BJRM	Summer	0.60 (0.15)	0.42 (0.03)	1.41 (0.27)	1.02 (0.18)	0.70 (0.19)	1.72 (0.36)
		Autumn	1.07 (0.14)	1.08 (0.09)	0.99 (0.05)	2.15 (0.24)	0.78 (0.09)	2.93 (0.33)
		Winter	1.18 (0.06)	1.43 (0.56)	0.90 (0.29)	2.61 (0.62)	0.86 (0.04)	3.47 (0.66)
		Spring	1.28 (0.02)	2.80 (0.10)	0.46 (0.02)	4.09 (0.09)	1.11 (0.08)	5.19 (0.16)
	BJRD	Summer	0.43 (0.02)	0.46 (0.04)	0.95 (0.10)	0.89 (0.03)	0.54 (0.05)	1.43 (0.08)
		Autumn	1.12 (0.07)	1.17 (0.19)	0.98 (0.21)	2.28 (0.17)	1.05 (0.15)	3.33 (0.31)
		Winter	0.94 (0.09)	0.94 (0.01)	1.00 (0.08)	1.87 (0.10)	0.54 (0.01)	2.42 (0.11)
		Spring	1.12 (0.08)	2.70 (1.31)	0.48 (0.21)	3.82 (1.23)	1.17 (0.08)	4.99 (1.31)
WJR	WJRU	Summer	0.14 (0.07)	0.16 (0.02)	0.85 (0.32)	0.30 (0.09)	0.27 (0.10)	0.57 (0.19)
		Autumn	0.73 (0.04)	0.46 (0.05)	1.62 (0.25)	1.19 (0.02)	0.40 (0.03)	1.58 (0.04)
		Winter	0.92 (0.07)	0.69 (0.16)	1.41 (0.43)	1.60 (0.16)	0.27 (0.05)	1.87 (0.21)
		Spring	1.07 (0.30)	1.16 (0.20)	0.91 (0.13)	2.23 (0.50)	0.46 (0.04)	2.70 (0.54)
	WJRM	Summer	0.09 (0.01)	0.11 (0.03)	0.83 (0.25)	0.19 (0.02)	0.13 (0.06)	0.32 (0.08)
		Autumn	0.63 (0.02)	0.56 (0.18)	1.20 (0.30)	1.18 (0.19)	0.37 (0.05)	1.56 (0.25)
		Winter	0.84 (0.15)	0.58 (0.11)	1.45 (0.02)	1.42 (0.25)	0.22 (0.08)	1.63 (0.33)
		Spring	0.37 (0.23)	0.87 (0.11)	0.41 (0.21)	1.25 (0.35)	0.36 (0.22)	1.60 (0.57)
	WJRD	Summer	0.17 (0.05)	0.13 (0.06)	1.37 (0.39)	0.29 (0.09)	0.15 (0.08)	0.44 (0.17)
		Autumn	0.44 (0.07)	0.59 (0.07)	0.75 (0.08)	1.03 (0.13)	0.41 (0.01)	1.44 (0.14)
		Winter	0.79 (0.02)	0.56 (0.03)	1.43 (0.07)	1.35 (0.05)	0.57 (0.15)	1.92 (0.20)
		Spring	0.48 (0.08)	0.72 (0.09)	0.67 (0.05)	1.20 (0.18)	0.20 (0.01)	1.40 (0.18)
Two-way ANOVA results								
River			***	***	**	***	***	***
Season			***	***	***	***	***	***
River × Season			ns	***	ns	***	***	***

Values are means (n = 3) with standard deviation (in parentheses).
**: $p < 0.01$; ***: $p < 0.001$; ns: not significant.
[a] BJRU: Bailongjiang River Upstream; BJRM: Bailongjiang River Midstream; BJRD: Bailongjiang River Downstream; WJRU: Wulongjiang River Upstream; WJRM: Wulongjiang River Midstream; WJRD: Wulongjiang River Downstream.

3.2. Relationships between precipitation, soil and foliar N variables

The concentrations of NO_3^--N were positively related to the concentrations of NH_4^+-N in BP ($p < 0.001$) and TP ($p < 0.001$; Fig. 3a). There were significant positive linear relationships between the DON concentrations and both the NH_4^+-N, NO_3^--N, DIN and TDN concentrations in BP and TP across the two rivers (all $p < 0.001$; Fig. 3b–e). The DIN, DON and TDN concentrations of TP were positively related to the DIN, DON and TDN concentrations of BP, respectively (both $p < 0.001$; Fig. 3f). Soil total nitrogen (TN) and $\delta^{15}N$ showed significant positive linear relationships with precipitation TDN (all $p < 0.05$; Fig. 4a and b). We also observed a significant positive linear relationship between the foliar $\delta^{15}N$ and precipitation TDN ($p = 0.04$; Fig. 4d) but not between the foliar TN and precipitation TDN ($p = 0.54$; Fig. 4c).

4. Discussion

All N forms in BP and TP showed large spatial and temporal variations. Significantly higher NH_4^+-N, NO_3^--N, DIN, DON and TDN concentrations in both BP and TP were observed in the more urbanized BJR than in the less urbanized WJR (Tables 1 and 2). This indicates that the higher urbanization level in the BJR forests has greatly increased N emissions from multiple anthropogenic sources, mostly domestic sources (e.g. sewage) as well as industrial, transportation, and energy production sources (i.e. fossil fuel combustion). This in turn has increased the concentrations of different N forms in precipitation over the BJR forests than over the WJR forests. Our results are similar to those reported in previous urban studies. For instance, a study by Lovett et al. (2000) in oak-dominated forests across an urbanization gradient in United States observed a significant decline in NH_4^+-N and NO_3^--N concentrations and fluxes with increasing distance from the city. Based on their study in subtropical forests of the Guangzhou metropolitan region in southern China, Huang et al. (2012) have reported substantially greater atmospheric inorganic N deposition in urban forests compared with suburban and rural forests. The two aforementioned studies have attributed their steep urban-rural N concentration and flux gradients to greater fossil fuel combustion-derived pollutant concentrations (e.g. NO_x gases) in the urban atmosphere than in the rural atmosphere. Another study in south-eastern Brazil found that as the distance from the coastal urban region increases, the bulk concentrations of all N forms including DON and TDN decreases, highlighting elevated anthropogenic N emissions originating predominantly from urban sewage and transportation sources (de Souza et al., 2015).

Table 2
Concentrations of ammonium-nitrogen (NH_4^+-N), nitrate-nitrogen (NO_3^--N), NH_4^+-N/NO_3^--N ratio, dissolved inorganic nitrogen (DIN), dissolved organic nitrogen (DON) and total dissolved nitrogen (TDN) in throughfall precipitation across seasons in forest stands of the Bailongjiang (BJR) and Wulongjiang (WJR) rivers.

River	[a]Stand	Season	NH_4^+-N (mg L^{-1})	NO_3^--N (mg L^{-1})	Ratio	DIN (mg L^{-1})	DON (mg L^{-1})	TDN (mg L^{-1})
BJR	BJRU	Summer	0.94 (0.54)	2.47 (0.71)	0.41 (0.28)	3.41 (0.79)	1.53 (0.34)	4.94 (1.09)
		Autumn	2.87 (0.72)	5.20 (0.90)	0.57 (0.18)	8.07 (1.17)	2.44 (0.81)	10.51 (1.97)
		Winter	2.18 (0.33)	3.92 (1.82)	0.61 (0.19)	6.10 (2.03)	1.26 (0.29)	7.35 (2.32)
		Spring	4.07 (1.15)	6.32 (1.83)	0.65 (0.10)	10.39 (2.83)	2.33 (0.67)	12.72 (3.50)
	BJRM	Summer	1.12 (0.11)	1.38 (0.05)	0.81 (0.05)	2.50 (0.15)	1.02 (0.29)	3.52 (0.44)
		Autumn	1.88 (0.22)	2.42 (0.58)	0.79 (0.11)	4.30 (0.77)	1.70 (0.46)	6.01 (1.23)
		Winter	1.56 (0.44)	3.25 (1.31)	0.51 (0.12)	4.81 (1.65)	1.56 (0.21)	6.38 (1.86)
		Spring	1.90 (0.88)	3.13 (1.30)	0.64 (0.24)	5.02 (2.04)	2.03 (0.69)	7.05 (2.71)
	BJRD	Summer	0.50 (0.06)	0.50 (0.14)	1.08 (0.45)	1.00 (0.09)	0.72 (0.12)	1.71 (0.21)
		Autumn	1.45 (0.07)	2.09 (0.31)	0.70 (0.07)	3.53 (0.37)	1.38 (0.19)	4.91 (0.55)
		Winter	1.21 (0.79)	1.98 (0.68)	0.59 (0.24)	3.19 (1.39)	1.05 (0.24)	4.24 (1.64)
		Spring	1.71 (0.52)	1.88 (0.58)	0.91 (0.07)	3.59 (1.08)	1.61 (0.64)	5.20 (1.72)
WJR	WJRU	Summer	1.60 (1.03)	0.41 (0.10)	3.61 (1.92)	2.01 (1.13)	1.03 (0.37)	3.04 (1.50)
		Autumn	2.02 (0.75)	2.01 (0.35)	0.99 (0.20)	4.03 (1.09)	1.24 (0.23)	5.27 (1.32)
		Winter	1.47 (0.30)	1.76 (0.37)	0.87 (0.28)	3.23 (0.45)	1.00 (0.37)	4.23 (0.81)
		Spring	1.31 (0.22)	1.37 (0.75)	1.10 (0.43)	2.67 (0.87)	1.23 (0.62)	3.90 (1.49)
	WJRM	Summer	0.25 (0.11)	0.42 (0.17)	0.59 (0.02)	0.67 (0.29)	0.41 (0.11)	1.08 (0.39)
		Autumn	1.15 (0.15)	1.36 (0.71)	1.04 (0.56)	2.51 (0.63)	0.74 (0.31)	3.25 (0.93)
		Winter	1.74 (0.20)	1.48 (0.35)	1.21 (0.19)	3.22 (0.53)	0.82 (0.24)	4.04 (0.77)
		Spring	1.46 (0.34)	2.52 (1.39)	0.74 (0.45)	3.98 (1.26)	0.66 (0.20)	4.64 (1.46)
	WJRD	Summer	0.58 (0.02)	0.54 (0.07)	1.10 (0.14)	1.12 (0.09)	0.42 (0.02)	1.54 (0.11)
		Autumn	0.62 (0.07)	0.63 (0.30)	1.17 (0.63)	1.25 (0.23)	0.77 (0.17)	2.02 (0.40)
		Winter	1.24 (0.13)	1.33 (0.34)	0.97 (0.21)	2.58 (0.40)	0.80 (0.04)	3.38 (0.44)
		Spring	1.27 (0.16)	1.36 (0.46)	0.99 (0.24)	2.62 (0.56)	0.36 (0.17)	2.98 (0.73)
Two-way ANOVA results								
River			**	***	**	***	***	***
Season			***	***	ns	***	**	***
River × Season			ns	ns	ns	ns	*	ns

Values are means (n = 3) with standard deviation (in parentheses).
*: $p < 0.05$; **: $p < 0.01$; ***: $p < 0.001$; ns: not significant.
[a] BJRU: Bailongjiang River Upstream; BJRM: Bailongjiang River Midstream; BJRD: Bailongjiang River Downstream; WJRU: Wulongjiang River Upstream; WJRM: Wulongjiang River Midstream; WJRD: Wulongjiang River Downstream.

There was a significant positive relationship between NO_3^--N and NH_4^+-N concentrations (Fig. 3a), indicating that they were both positively influenced by the increase in urbanization-induced anthropogenic N sources. Specifically, the NH_4^+-N concentrations may be influenced by NH_3 volatilization from multiple sources including fertilizers, excretory wastes and sewage whereas the NO_3^--N concentrations might be dependent on NO_x emissions from fossil fuel combustion in industrial, transportation as well as energy production sources. Positive linear relationships were noticed between the DON concentrations and both the NH_4^+-N, NO_3^--N, DIN and TDN concentrations (Fig. 3b–e). These relationships suggest that these N forms might have originated from similar anthropogenic N sources. Our results are consistent with those from previous studies (e.g. Zhang et al., 2011; Zhang et al., 2012; Li et al., 2012).

The contributions of NO_3^--N to the TDN exceeded those of NH_4^+-N in the BJR forests (Fig. 2), suggesting that NO_3^--N is the dominant N pollutant in the precipitation falling on the forests along this river. Because of its traditional agriculture, which is the largest source of ammonia (NH_3) emissions into the atmosphere, China has been experiencing atmospheric deposition with much higher NH_4^+-N than NO_3^--N (e.g. Chen et al., 2004; Zhang et al., 2007; Fang et al., 2009, 2011; Zhan et al., 2015). For example, Chen et al. (2004) reported that agricultural-related NH_4^+-N accounted for about 70% of the atmospherically deposited N in subtropical forested catchments in south China. Even in urban areas, NH_4^+-N has usually been found to be the dominant N form in atmospheric N deposition, mostly due to its elevated emissions from excretory wastes and sewage (Liu et al., 2008; Xiao et al., 2010).

However, the DIN composition in atmospheric N deposition across the BJR forests changes our previous perception that agricultural-related NH_4^+-N often dominate fossil fuel combustion-related NO_3^--N in atmospheric N deposition across China. Our findings revealed that urbanization-induced anthropogenic activities, especially those related to fossil fuel combustion, are increasing more sharply along the BJR forests, resulting in greater NO_3^--N emissions into the atmosphere and its growing contributions to the TDN as evidenced by the higher NO_3^--N than NH_4^+-N in atmospheric N deposition. These findings were further supported by the NH_4^+-N/NO_3^--N ratio, which was less than 1, suggesting that precipitation N concentrations over the BJR forests are primarily influenced by fossil fuel combustion-related activities. As in our study, NO_3^--N accounted for 50–63% of the total inorganic N deposition between 2001 and 2006 in Shenzhen city (Huang et al., 2013). Similar patterns were observed in a rapidly urbanizing Pearl River Delta Region in southern China, where 53.5–79.1% of the total wet inorganic N inputs were in NO_3^--N form (Huang et al., 2015).

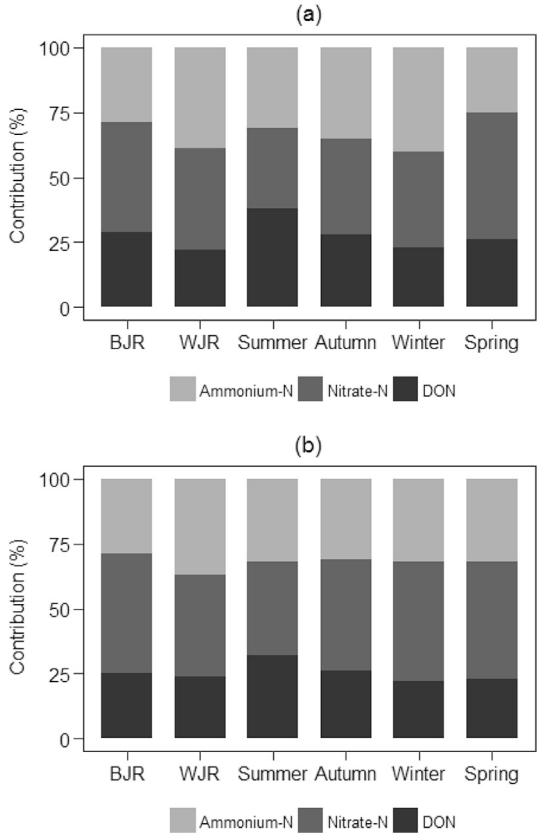

Fig. 2. Contributions of ammonium-nitrogen (NH_4^+-N), nitrate-nitrogen (NO_3^--N) and dissolved organic nitrogen (DON) to the total dissolved nitrogen (TDN) in bulk (a) and throughfall (b) across seasons and rivers. BJR: Bailongjiang River; WJR: Wulongjiang River.

In contrast, the contributions of NO_3^--N to the TDN were almost equal to those of NH_4^+-N in the WJR forests, which is supported by the 1.14 NH_4^+-N/NO_3^--N ratio, indicating that agricultural and industrial activities collectively influence atmospheric N emissions and deposition in forest ecosystems along this river. Similarly, the NH_4^+-N/NO_3^--N ratio was on average 1.23 in the Yangtze River Delta Region (Zhao et al., 2009) and 1.22 in Chinese terrestrial ecosystems across eight ecological regions (Zhu et al., 2015), suggesting increasing importance of atmospheric NO_3^--N deposition and that agricultural and industrial N emissions were the two major sources of atmospheric N deposition.

As one recent review-based study insisted that the concentrations of NH_x are generally higher than those of NO_x in urban N deposition not only across China but also across the world (Decina et al., 2019) and another most recent research-based study that still revealed enhanced NH_x concentrations in Beijing urban atmosphere (Zhang et al., 2020), it is therefore worth highlighting that the influence of urbanization on the shifts in dominance between reduced and oxidized N forms in urban precipitation is a topic that is perhaps still open to scientific debate.

Furthermore, we found substantial contributions of DON to the TDN in precipitation along both rivers (22–29%; Fig. 2), which is consistent with the contributions reported in previous urban research around the world e.g. coastal urban region in southeastern Brazil (32–56%; de Souza et al., 2015), Keelung coastal city in southern East China Sea (37%; Chen et al., 2015), peri-urban evergreen holm oak forests in Spain (38–40%; Izquieta-Rojano et al., 2016) and greater Boston area in United States (35–38%; Decina et al., 2018). Our results indicate that the DON is a significant component of N deposited in precipitation particularly in N-polluted urban atmospheres. However, the contributions of DON to the TDN were more pronounced in the more urbanized BJR forests compared with the less urbanized WJR forests (Fig. 2), highlighting increase in DON emission sources with decreasing distance to the city center. Similar patterns were found by de Souza et al. (2015) in southeastern Brazil, where the contributions of DON to the TDN were greater in the coastal urban site (56%) than in the coastal peri-urban site (32%).

Moreover, the concentrations of most N forms in summer were generally lower relative to those in other seasons, in particular winter and spring seasons (Tables 1 and 2). This can be attributed to the dilution effect of precipitation on the concentrations of different N forms during the summer, where the precipitation amount was greater than in the three other seasons (Fig. 1). This phenomenon has been documented in previous atmospheric N deposition studies (Zhao et al., 2009; Yu et al., 2011; Li et al., 2012). This also suggests higher heating-related N emissions in the winter owing to excessive cold, which

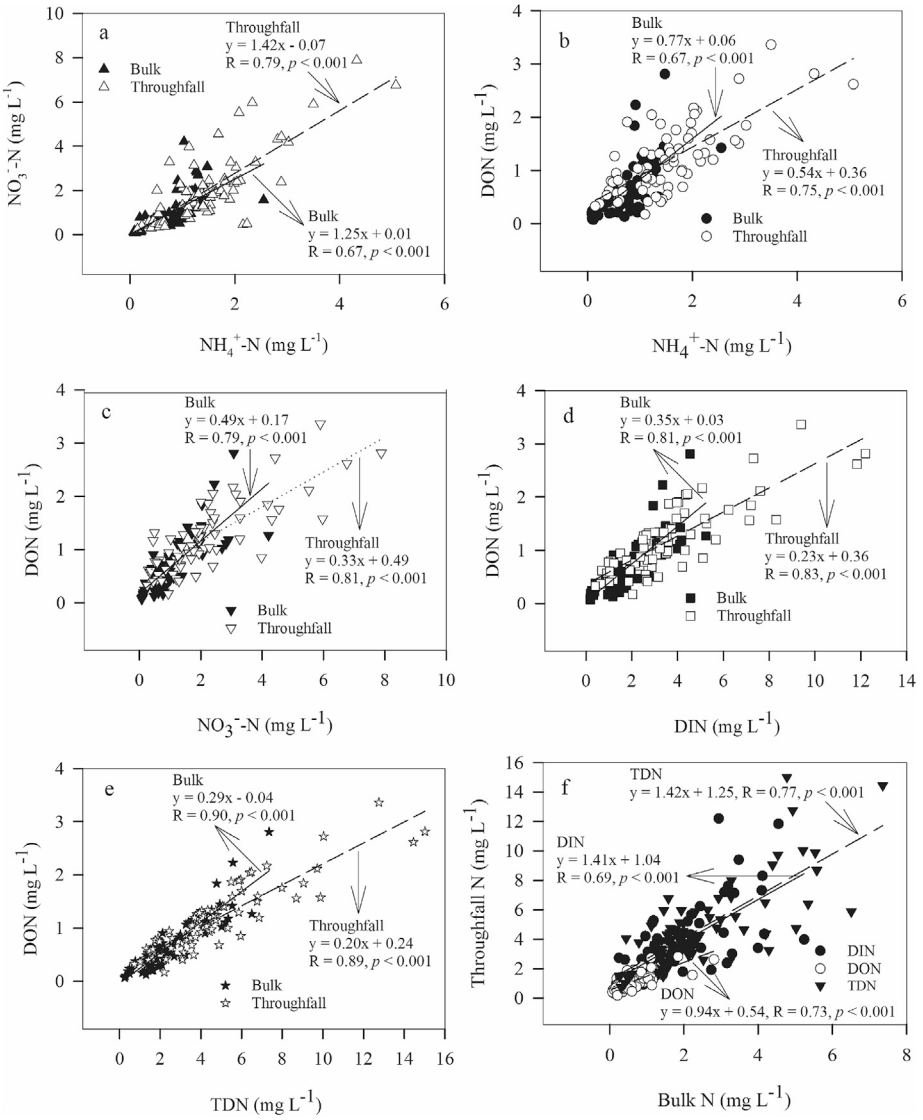

Fig. 3. Linear relationships between nitrate-nitrogen and ammonium-nitrogen (**a**), and between dissolved organic nitrogen and ammonium-nitrogen (**b**), nitrate-nitrogen (**c**), dissolved inorganic nitrogen (**d**) and total dissolved nitrogen (**e**), as well as between throughfall and bulk N forms (**f**) across the study rivers.

is supported by elevated N concentrations in this season and later in the spring. Zhan et al. (2015) also found enhanced throughfall DIN deposition in Mao'ershan mixed conifer-broadleaved forests in China during the colder winter months, which they attributed to an increase in fossil fuel combustion-derived N emissions, as driven primarily by an increase in winter heating activities.

The NO_3^--N contributions to the TDN were usually greater compared with the NH_4^+-N contributions (Fig. 2), indicating the dominance of NOx-N emissions from fossil fuel combustion-related activities over NHx-N emissions from agricultural-related activities in precipitation falling across the seasons. In addition, lower DON contributions to the TDN were noticed in the autumn, winter and spring than in the summer. The results may suggest elevated presence of less soluble atmospheric organic nitrogen compounds including alkyl nitrates, peroxyacetyl nitrate and alkyl cyanides (Cornell et al., 2003). The concentrations of throughfall N forms were considerably greater compared with the concentrations of bulk N forms (Tables 1 and 2), indicating higher dry N deposition wash-off on canopy surfaces and foliar N leaching during precipitation. Similar findings were also found across urban forests in Lithuania (Juknys et al., 2007), China (Fang et al., 2011) and United States (Decina et al., 2018). Also, the DIN, DON and TDN concentrations in TP were positively related to those in BP (Fig. 3f). These relationships suggest that although TP and BP nitrogen concentrations differed significantly, they both had similar spatial patterns, with their values increasing from the less urbanized WJR towards the more urbanized BJR. Our findings agree with the results of Zhan and colleagues across eight forest ecosystems in China, where there was a significant positive relationship

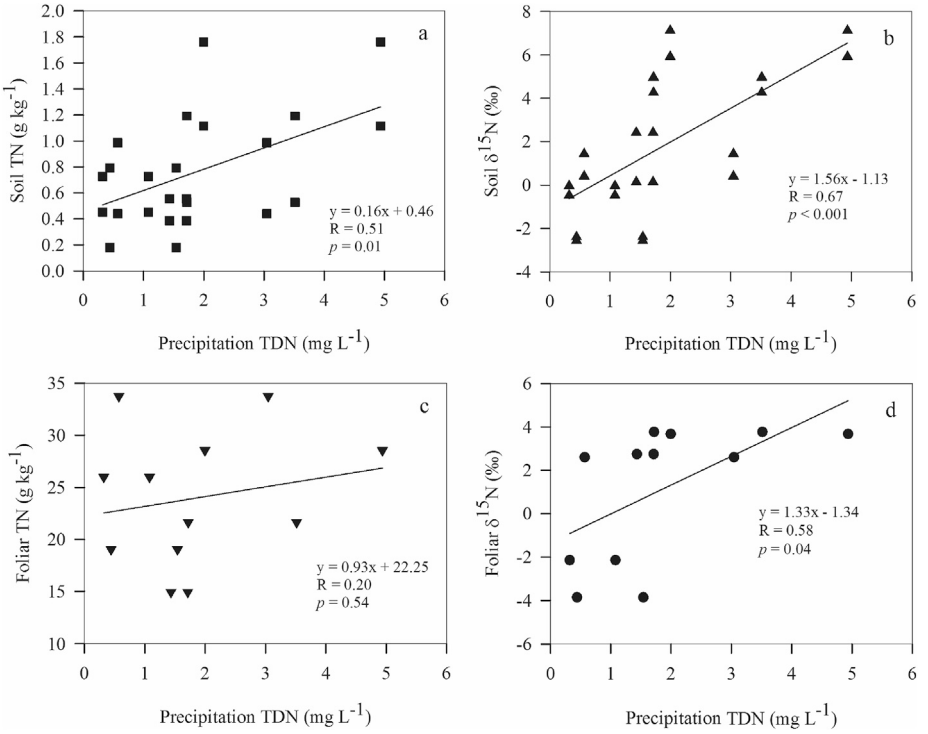

Fig. 4. Linear relationships between precipitation total dissolved nitrogen and soil total N (**a**), soil $\delta^{15}N$ (**b**), foliar total N (**c**) and foliar $\delta^{15}N$ (**d**) across the study sites in Fuzhou coastal city. **Note**: soil and foliar data were taken from the previous study (Mgelwa et al., 2019a).

between bulk and throughfall DIN deposition (Zhan et al., 2015). The contributions of NO_3^--N to the TDN in TP tended to be higher compared to those in BP, thereby highlighting the vital contributions of foliar NO_3^--N leaching as well as microbial nitrification and dry deposited NO_3^--N on forest canopies to the throughfall N concentrations.

To understand the consequences of atmospheric N deposition on soil and plant N availability, we correlated precipitation TDN data from our current study with soil and foliar TN and $\delta^{15}N$ data from our previous study (Mgelwa et al., 2019a). We found a significant positive relationship between soil TN concentrations and precipitation TDN concentrations (Fig. 4a), indicating that soil N availability in forest ecosystems across the two rivers is influenced by atmospheric N deposition and that its observed enhancement could potentially lead to soil N saturation. The atmospheric N deposition has been reported to enhance soil N availability (Tian et al., 2018). Soil net N mineralization and net nitrification rates were found to increase with increasing soil N availability (Mgelwa et al., 2019a). Consequently, the ^{15}N-depleted NH_4^+-N (mineralization product) and ^{15}N-depleted NO_3^--N (nitrification product) might have been preferentially lost, resulting in significant enrichment of ^{15}N in the residual soil N pools. Therefore, the ^{15}N enrichment in soils along the BJR and WJR urban forests as reported in our previous study could be attributed to the atmospherically deposited N, which is supported by their significant positive linear relationship (Fig. 4b). The foliar $\delta^{15}N$ values were positively related to the precipitation TDN values (Fig. 4d). Because the foliar $\delta^{15}N$ values often reflect the source $\delta^{15}N$ values, the aforesaid relationship may suggest plant N uptake of the atmospherically deposited nitrogen that could represent an important N source in urban forests along the two rivers. It is therefore worth examining the precipitation $\delta^{15}N$ values in our future research to better understand their influence on foliar $\delta^{15}N$ patterns across urban forest ecosystems. The lack of relationship between foliar TN concentrations and precipitation TDN concentrations in the current study was slightly surprising (Fig. 4c) as elevated N availability was expected to enhance plant N uptake (Jones and Power, 2012). Our finding indicates that the interaction between foliar TN and precipitation TDN is complex and it requires global attention.

In conclusion, our findings clearly showed that forests along the more urbanized BJR are receiving elevated precipitation N concentrations originating primarily from fossil fuel combustion and that serious N cycle perturbations may occur in these forests if urgent measures are not taken. The increase in NO_3^--N dominance from the less urbanized WJR forests to the more urbanized BJR forests suggests that rapid urbanization in developing countries is changing the dominance in atmospheric inorganic N deposition from NH_4^+-N dominance to NO_3^--N dominance. The substantial DON contributions to the TDN in our study indicate that DON is gradually becoming a significant component of N deposited in precipitation especially in N-polluted urban atmospheres and that it should be incorporated in atmospheric N deposition studies as well as in various initiatives dealing with N pollution control and management. Since our results suggested that heating activities are the main sources of atmospheric N pollutants during the colder winter months, using eco-friendly heating technologies that emit less

N pollutants to the atmosphere could be an essential approach to controlling N pollution in urban ecosystems. Moreover, as urban tree canopies were found to enhance precipitation N concentrations, urban trees should be planted above permeable surfaces where precipitation can easily infiltrate soils to reduce N loads in urban watersheds. Additionally, forest ecosystem N availability responded positively to precipitation N concentrations, highlighting that elevated N deposition can have profound, long-term forest N saturation and N biogeochemical cycle consequences. This study is among the few documenting the concentrations of dissolved inorganic and organic nitrogen in both bulk and throughfall precipitation. The findings of this study are useful to urban planners and decision-makers globally in their efforts to look for suitable N pollution abatement strategies and to look at the effects of increasing atmospheric N deposition on forest ecosystems. To further support their efforts, our future research should include novel methods (e.g. stable isotopes) to identify various N sources in atmospheric deposition across urban forests.

Declaration of competing interest

We declare that we have no conflicts of interest.

Acknowledgments

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