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Epilithic moss as a bio-monitor of atmospheric N deposition in South China

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[1] To assess the utility of epilithic mosses as bio-monitors of atmospheric N deposition in Southern China, single and multiple moss species were collected in forested, agricultural, suburban, urban, and sewage landfill habitats, and then analyzed for N tissue content and δ^{15}N values. About 83% of all the moss samples showed an N content of >2.0% and significant differences were only found between forests and the other four kinds of areas. But no significant differences were found between single and multiple moss species at each area. Six previously reported relationships between N deposition and N contents of single moss species and four of multiple moss species were assessed to be used for estimation of N deposition. In the selected areas of South China, the average of N deposition estimated using the equations of single and multiple moss species was 30.8 to 41.6 kg N ha^{-1} yr^{-1} and 26.0 to 34.9 kg N ha^{-1} yr^{-1}, respectively. And the former showed much larger standard deviation of N deposition than the latter, suggesting that the equations of multiple moss species might be preferred to be used for estimation of N deposition. There were significant differences in moss δ^{15}N values in different kinds of areas except between urban areas and sewage landfills, where mosses were most ^{15}N-depleted (−10‰ to −2‰, reflecting sewage-derived NH_y). For both single and multiple moss species, those in suburban areas were between −5‰ to −1‰, reflecting the mixing of sewage-derived and soil-derived NH_y. The δ^{15}N values of mosses in forested (−3‰ to +3‰) and agricultural areas (−1‰ to +1‰) indicated soil-derived NH_y and chemical fertilizer.

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1. Introduction

[2] Over the past half century there has been a substantial increase in atmospheric deposition of fixed N as a consequence of the enhanced use of nitrogenous fertilizers and fossil fuel consumption, and this increase is expected to be particularly intense in Asia and Africa [*Galloway et al.*, 1995]. The enhanced N inputs which lead to N saturation in many ecosystems has various negative effects, including enhanced leaching of nitrate and soil nutrients [*Aber et al.*, 1998], soil acidification of soil and water [*Vitousek et al.*, 1997], decreased mycorrhizal root symbiosis [*Lilleskov et al.*, 2002], ecosystem eutrophication followed by losses of biodiversity and decline of forest ecosystems [*Bobbink et al.*, 1998].

[3] In evaluating the role of atmospheric N deposition on terrestrial ecosystems, it is critical to investigate the levels of both wet and dry deposition. Accurate estimation of the annual mean total deposition is usually difficult because nitrogen can be deposited on a surface in different forms and it appears in the lower atmosphere in a variety of N compounds. And therefore it is usually restricted to short-term field campaigns at selected localities [*Xiang et al.*, 2006; *Fan et al.*, 2007]. Although N emissions have significantly increased in China [*Xiao and Liu*, 2002], little is known about N deposition in South China.

[4] Carpet-forming ectohydric mosses have successfully been used as biomonitors of atmospheric N deposition in the past decade [*Pitcairn et al.*, 2006; *Salemaa et al.*, 2008; *Zechmeister et al.*, 2008]. These mosses obtained N directly from wet and dry deposition with little uptake from the substrate. Nitrogen contents in moss tissue provide a surrogate, time-integrated measurement of N deposition from the atmosphere to terrestrial systems [e.g., *Salemaa et al.*, 2008].

[5] Many previous studies indicated a possible link between N deposition and N contents in moss tissue, as summarized in Table 1. For instance, *Thöni et al.* [2008] found a correlation between moss tissue N content and measured N deposition in Switzerland. *Harmens et al.* [2005] also reported a strong linear relationship between the total N contents in mosses and EMEP modeled atmospheric N deposition rates. Recently, we integrated some previously reported data as a good linear equation for calculating atmospheric N deposition in the Yangtze River drainage basin [*Xiao et al.*, 2010a]. Although tissue N contents increased linearly with increasing

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Table 1. A Summary of the Relationship Between Atmospheric N Deposition (x) and Tissue N Contents in Mosses (y)

	Moss Species	Equations[a]	R^2	p Value	Regions	References
		Single Moss Species				
A	*Dicranum polysetum*	$y = 0.627 + 0.0388x$	nd	<0.000	Finland	*Salemaa et al.* [2008]
B	*Hylocomium splendens*	$y = 0.6 + 0.04x$	0.39	<0.001	England	*Hicks et al.* [2000]
C	*Hylocomium splendens*	$y = 0.725 + 0.0494x$	nd	<0.000	Finland	*Salemaa et al.* [2008]
D	*Pleurozium schreberi*	$y = 0.678 + 0.0552x$	nd	<0.000	Finland	*Salemaa et al.* [2008]
E	*Scleropodium purum*	$y = 0.551 + 0.061x$	0.341	<0.001	Germany	*Solga et al.* [2005]
F	*Pleurozium schreberi*	$y = 0.583 + 0.066x$	0.429	<0.001	Germany	*Solga et al.* [2005]
		Mixture of Several Moss Species				
G	mixture of mosses[b]	$y = 0.73 + 0.052x$	0.70	<0.001	China	*Xiao et al.* [2010a]
H	Pleurocarpous mosses	$y = 0.70 + 0.055x$	0.91	nd	Switzerland	*Thöni et al.* [2008]
I	mixture of *Pleurozium schreberi, Hypnum cupressiforme and Scleropodium purum*	$y = 0.53 + 0.08x$	0.71	nd	Scandinavian	*Harmens et al.* [2005]
J	mixture of *Sphagnum* mosses	$y = 2.5\ln(x/1000) + 11$	0.85	nd	11 European counties	*Bragazza et al.* [2005]

[a] The units of tissue N contents and atmospheric N deposition are % and kg N ha^{-1} yr^{-1}, respectively; nd, no data.
[b] Integrated from the data reported by *Pitcairn et al.* [1995, 2001, 2002], *Bragazza et al.* [2005] and *Solga et al.* [2005].

N deposition for most species (Table 1), they showed different outcomes under comparable conditions according to their different strategies regarding N uptake [*Pitcairn et al.*, 1995]. Moss species reached maximum N contents at different external N deposition levels. For example, *Dicranum* needed a longer N exposure to reach the maximum tissue content than *Pleurozium* and *Hylocomium* [*Salemaa et al.*, 2008]. Therefore, the reported correlation equations are usually species-specific. But most of the species used by those authors are not common enough to be used for monitoring in other regions. Therefore, in order to estimate N deposition in a new region, researchers need to assess the potential of different moss species for using tissue N contents as bio-monitors for N deposition.

[6] Stable nitrogen isotopic analysis has become a valuable tool in atmospheric environment research [e.g., *Heaton*, 1986; *Xiao and Liu*, 2002]. For instance, agricultural sources of N pollution as a result of strong use of chemical N fertilizer may contain an isotopically distinct signal, which may be distinguished from background values derived primarily from soils [*Moore*, 1974]. Nitrogen isotopes in mosses have also been used successfully to investigate the importance of different sources of N deposition [*Pearson et al.*, 2000; *Solga et al.*, 2005; *Zechmeister et al.*, 2008] because isotopic fractionation during N uptake by mosses has been assumed to be absent or very low [*Bragazza et al.*, 2005]. Moss $\delta^{15}N$ values in some cities and forests in the Yangtze River drainage basin (China) were found in distinctly different ranges and showed that the main N sources in most of these cities were excretory wastes and those in forests were soil emissions [*Xiao et al.*, 2010a]. A study in three areas of South China indicated that $\delta^{15}N$ signatures of mosses reflected those of nitrogen deposited from the atmosphere at some urban, rural and forested sites [*Xiao et al.*, 2010b].

[7] In some urban and forested areas in South China, we have determined the tissue N contents and ^{15}N natural abundance in epilithic mosses, and atmospheric N deposition was also estimated using an integrated equation [*Xiao et al.*, 2010a]. In this study, tissue N contents and $\delta^{15}N$ values of mosses in more areas such as suburban and agricultural areas, and some near waste landfill were analyzed. The aim of this paper was (a) to gather moss N content data in different kinds of areas and to use these data to predict N deposition rates based on previously reported relationships; (b) to investigate which one between single and multiple moss species was the better bio-monitor of atmospheric N deposition, and (c) to use ^{15}N signals in mosses as an indicator of N sources in these areas.

2. Materials and Methods

2.1. Sampling Methods

[8] In 2006 to 2009, two kinds of moss samples, one moss species (*Haplocladium microphyllum* (Hedw.)) and multiple moss species (including *Haplocladium microphyllum* (Hedw.), *Pleurozium schreberi* and *Hylocomium splendens*), were collected, respectively, in sixteen urban areas (including Lhasa), eight forested areas, twelve agricultural areas, two waste landfills and five suburban areas in South China (Figure 1). Urban mosses were mainly collected around parks or hills. Sampling sites in other areas were selected to be located in open habitats like heaths or clearings (<100 m^2)

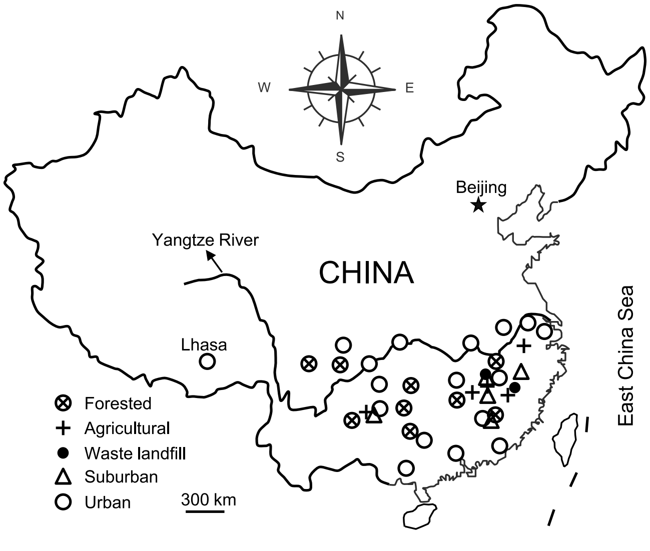

Figure 1. Map of China showing the sampling sites in South China. Some sampling sites are cited from *Xiao et al.* [2010a].

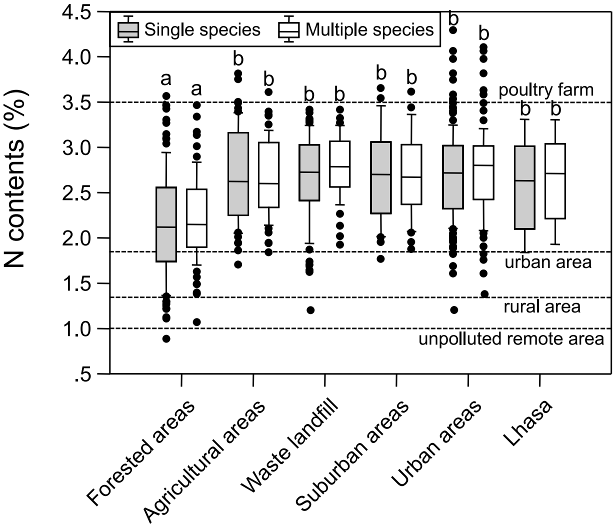

Figure 2. Moss tissue N contents in South China. Box plots display the 10th, 25th, 50th, 75th, and 90th percentiles as solid lines. Symbols indicate 5th and 95th percentiles. Data from different sampling sites that do not share a letter below the bars are significantly different at $p < 0.05$. The data at poultry farm, urban, rural and unpolluted remote areas are cited from *Pitcairn et al.* [1995, 2003] and *Pearson et al.* [2000].

and at least 500 m away from main roads and at least 100 m away from other roads or houses. All mosses were obtained from natural rocks without canopies or overhanging vegetation ensuring not influenced by throughfall N compounds and soil N. Sampling was performed only at those sites above ground level to avoid surface water splashes. Sites possibly disturbed by local point or non-point sources were also avoided. We collected 5–10 subsamples from different populations at each site and homogenized them into one representative sample. Only green, healthy samples were taken, avoiding yellow or dark samples.

2.2. Sample Preparation and Chemical Analyses

[9] Fresh mosses were dried in the sun quickly after sampling and then stored in cleaned plastic bags en route to the laboratory. Using the treatment method by *Liu et al.* [2007], samples were gently rinsed with 1.5 mol L^{-1} HCl solution, then sonicated and washed with deionized water for several times until no N (NH_4^+ and NO_3^-) was detected in the washed water (spectrophotometry, the limit of detection was <0.005 mg L^{-1}). The main purpose of this washing procedure was to remove adsorbed pollutants. All samples were dried in a vacuum oven at 70°C and re-dried after being ground separately in liquid nitrogen into fine powders using a mortar and pestle.

[10] Nitrogen contents of mosses were analyzed by an elemental analyzer (Model PE-2400 II, USA). The analytical precision of N was checked by analyzing cystine standard material (11.67%, N141–0324, provided by Perkin Elmer). Nitrogen contents (average ± SD) of the standard material were measured to be 11.7 ± 0.1% (n = 10). And a sample of this standard material was analyzed after each set of eight moss samples.

[11] After combustion at 850 °C and high purification with liquid nitrogen, nitrogen isotope ratios were determined on a Finnigan MAT 252 mass spectrometer. High purity N_2 reference gas was run with each analysis. Three to five replicate measurements per sample were carried out, and values were presented as the average of these measurements. Analysis of potassium nitrate standard (MOR2386–01) provided by Shoko Co., Ltd., Tokyo, Japan (+1.9‰) gave a mean (±SD) $\delta^{15}N_{air}$ value of 1.9 ± 0.2‰ (n = 5).

2.3. Statistical Analysis

[12] Statistical analysis was conducted using SPSS 11.5 statistical program, and graphs were mainly created with SigmaPlot 2000 software (both SPSS Science, Chicago, USA). Correlation analysis was carried out by calculating the Pearson product–moment correlation coefficient (r). Differences in contents and isotopic values between areas were tested for significance by using a one-way analysis of variance (ANOVA) procedure, and least significant differences (LSD) were used by a Tukey-HSD test to compare significant differences (at the 95% confidence level). Differences were considered significant at $P < 0.05$.

3. Results

3.1. Moss N Contents

[13] Tissue N contents of epilithic mosses in South China were in a wide range of 0.87% to 4.28%, with a mean of 2.55 ± 0.56% for single species and 2.57 ± 0.47% for multiple species, respectively. About 83% of all the moss samples had a tissue N content of >2.0%. Even in the eight forested areas, the percentage of >2.0% moss samples reached about 58%.

[14] As shown in Figure 2, tissue N contents of epilithic mosses of single and multiple species in the eight forested areas averaged 2.16% and 2.17%, respectively, significantly lower than those in other five selected areas ($p < 0.05$). The mean N contents in urban and agricultural mosses were very similar. Epilithic mosses of single and multiple species collected near two waste landfills showed slightly lower mean N contents (2.65% and 2.66%) than those growing in urban (2.69% and 2.73%) and agricultural areas (2.70% and 2.68%), while higher than those in suburban areas (2.60% and 2.58%). The moss N contents in Lhasa, a remote city in Tibet, also showed a higher mean value (2.57 ± 0.51%) as compared to those in forested areas. However, tissue N contents of epilithic mosses among agricultural, suburban and urban areas, waste landfill and even Lhasa were not significantly different ($p > 0.05$).

3.2. Atmospheric N Deposition

[15] Using all the previously reported relationship between tissue N contents in mosses and atmospheric N deposition (Table 1), we estimated the atmospheric N deposition in South China. The estimated results were shown in Figure 3.

[16] Among the six equations of single equations (Figure 3a), that using that via *Dicranum* reported by *Salemaa et al.* [2008] could get the highest level of atmospheric N deposition (line A) while using that by *Solga et al.* [2005] showed the lowest level (line F). For instance, in forested areas of South China, mean level of atmospheric N deposition was estimated to be about 39.5 kg N ha^{-1} yr^{-1} when the

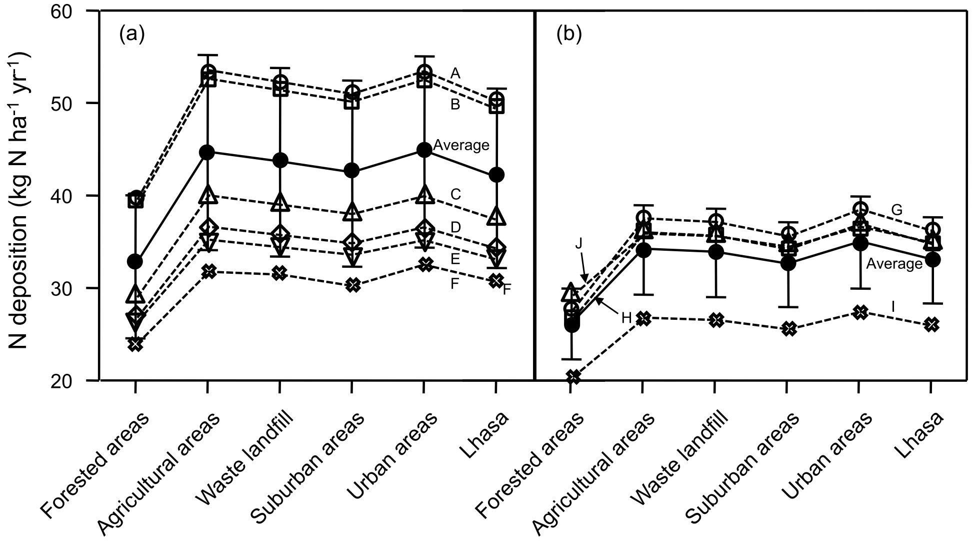

Figure 3. The distribution patterns of atmospheric N deposition in South China estimated using previously reported relationship as listed in Table 1. (a) Single moss species. (b) Multiple moss species.

A equation (Table 1) was used. The value was >1.5 times high than that estimated using line F (24.0 kg N ha^{-1} yr^{-1}).

[17] For the four equations of multiple species (Figure 3a), those estimated using lines G, H and J were very similar and much higher than those using line I. As compared to that estimated using equations of single mosses, N deposition estimated using different equations of multiple moss species showed a much narrower range at each area. But those estimated using lines C, D, E, F (Figure 3a) were similar to those estimated using lines G, H, J (Figure 3b). In the forested and agricultural areas, waste landfills, suburban and urban areas and Lhasa in South China, the average of N deposition estimated using the six equations of single moss species was estimated about 30.8, 41.6, 40.6, 39.6, 41.6 and 39.1 kg N ha^{-1} yr^{-1}, respectively, while that estimated using the four equations of multiple moss species was 26.0, 34.1, 33.7, 32.5, 34.9, 32.9 kg N ha^{-1} yr^{-1}, respectively.

3.3. ^{15}N Natural Abundance in Mosses

[18] Most of the epilithic mosses collected near waste landfill and in suburban and urban areas showed negative $\delta^{15}N$ values while those collected at Lhasa city (Tibet) showed positive $\delta^{15}N$ values (Figure 4), with a mean of +4.2 ± 2.5‰ and +3.1 ± 2.3‰ for single and multiple moss species, respectively. In urban areas and near waste landfill, more than 67% of the samples were found to be distributed in the range of −10‰ to −3‰ (more negative) and in suburban areas about 75% of the samples were between −5‰ and −1‰ (less negative). More than 78% of the samples collected in the forested areas were in the range of −3‰ to +3‰. And about 94% of the agricultural moss samples showed $\delta^{15}N$ values of ~0‰ (between −1‰ and +1‰).

[19] The between-area differences of moss $\delta^{15}N$ values were significant ($p < 0.05$) except that between waste landfill and urban areas (Figure 5). For both single and multiple moss species, the significant between-area differences were suggestive of some different N sources while mosses near waste landfill and in urban areas were exposed to N deposition with the same N sources.

4. Discussion

4.1. Moss N Contents Indicating Atmospheric N Deposition

[20] Due to the lack of site-based N deposition data, only a few studies have established the quantitative relation between N contents in natural growing mosses and the corresponding atmospheric N deposition (Table 1), and thus up until now they have rarely been used in regional N deposition surveys. In this study, levels of atmospheric N deposition estimated using the G equation (slope = 0.052) and the N contents of multiple moss species were 27.5 kg N ha^{-1} yr^{-1} in forested areas, very similar to the measured value (24 to 28 kg N ha^{-1} yr^{-1}) at a site in Caijiatang Forest, South China [*Xiang et al.*, 2006]. In Guiyang city, N deposition estimated using the G equation was about 30.18 kg N ha^{-1} yr^{-1}, which approximated to the measured mean value (31 kg N ha^{-1} yr^{-1}, H.-Y. Xiao et al., unpublished data, 2010). These maybe suggested a high accuracy in assessing N deposition level when using multiple moss species.

4.2. ^{15}N Natural Abundance in Mosses Indicating Sources of Atmospheric N Deposition

[21] In the past ten years more and more studies pointed to the fact that the consideration of $^{15}N/^{14}N$ ratio of mosses allowed differentiations of anthropogenic emission sources [*Bragazza et al.*, 2005; *Solga et al.*, 2005]. In the Netherlands, Denmark, Switzerland, and Italy, some very negative $\delta^{15}N$ values of mosses appeared reflected a major role of local ammonia emitted from excretory wastes [*Asman et al.*, 1998]. Studies by *Pearson et al.* [2000] in the London area and *Gerdol et al.* [2002] in northern Italy have found that moss $\delta^{15}N$ values can effectively decipher atmospheric N sources from urban traffic NO_x (relatively positive signature) and from rural animal NH_y (relatively negative signature).

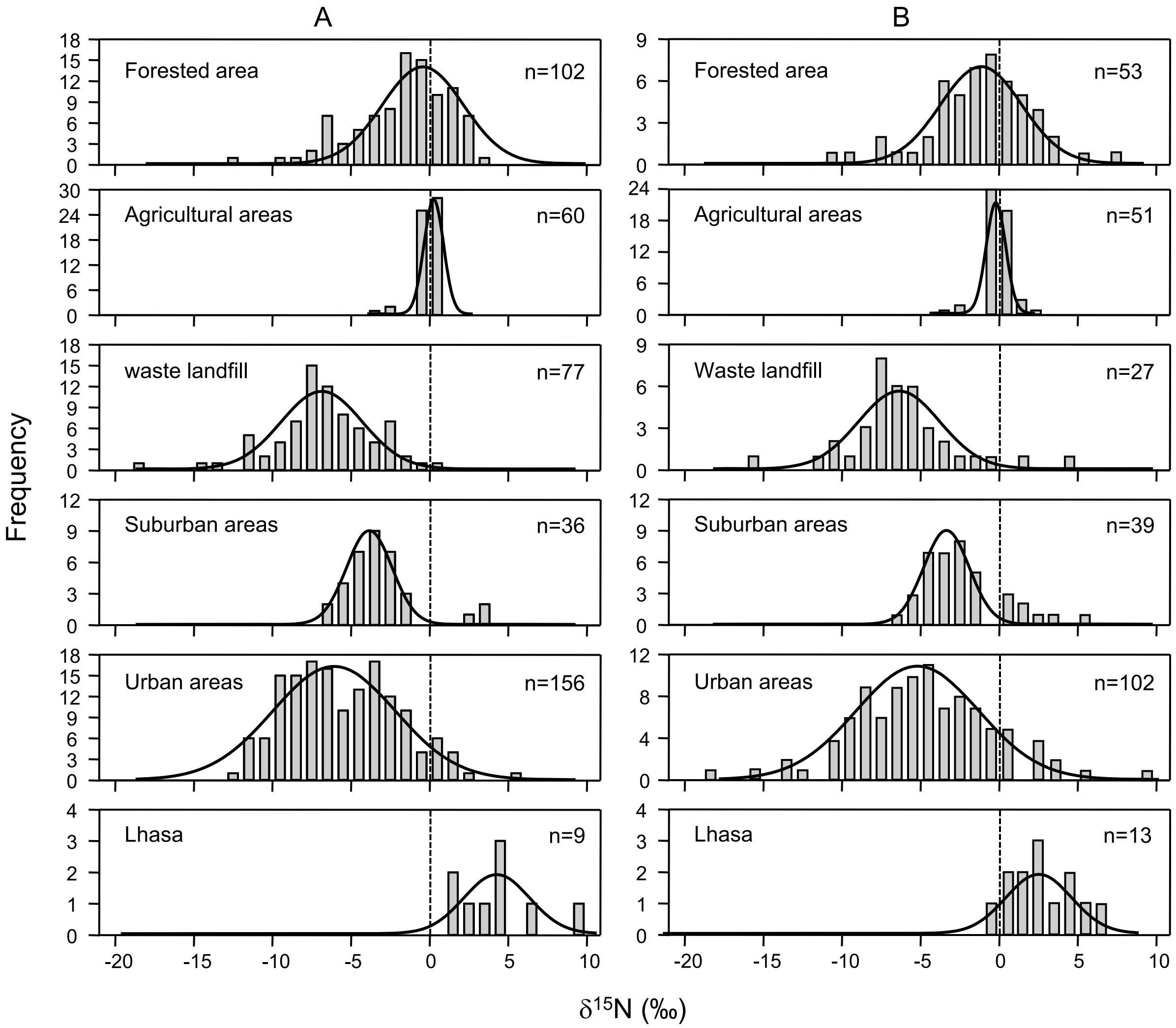

Figure 4. Frequency histograms of δ^{15}N values of (a) single species and (b) multiple species in South China.

Recently, more positive values were also observed in urban mosses than in forested mosses in Yangtze River drainage basin because urban mosses were exposed to sewage-derived NH_y (more ^{15}N-depleted) while forested mosses to soil-derived NH_y (less ^{15}N-depleted) [*Xiao et al.*, 2010a]. In summary, moss δ^{15}N signatures were controlled by the following three major factors as discussed below.

[22] There is wide agreement that oxidized (NO_x) and reduced (NH_y) N compounds have different δ^{15}N values [*Freyer*, 1978; *Garten*, 1992; *Heaton et al.*, 1997]. The latter (NH_y) is usually strongly depleted in ^{15}N. For this reason, a plant that takes up N predominantly in the form of NH_y will show negative δ^{15}N values in its tissue. NO_x, on the other hand, originates mostly from the combustion of fossil fuels such as coals and oils. It is almost always enriched in ^{15}N [*Heaton*, 1990; *Freyer*, 1991] and therefore, if a plant prefers to take up this as the main N species (e.g., mosses in Lhasa city), the analysis of its tissue will yield positive δ^{15}N values (Figure 5). In Guiyang city, our previous studies [*Liu et al.*, 2008] also showed that negative δ^{15}N signatures of urban mosses were rather closer to the $\delta^{15}NH_4^+$ value (-12.2 ± 6.7‰) than to the $\delta^{15}NO_3^-$ value ($+2.0 \pm 4.4$‰) in rainwater [*Xiao and Liu*, 2002], suggesting that urban mosses were more influenced by NH_y than by NO_x [*Xiao and Liu*, 2002, 2004]. The effect of relative contributions of NH_y to NO_x on moss δ^{15}N values could be also seen from the negative correlation between moss δ^{15}N values and the ratios of NH_y–N/NO_x–N in N deposition [*Solga et al.*, 2005; *Bragazza et al.*, 2005]. This meant that the higher the NH_y–N/NO_x–N ratios in N deposition, the more negative the moss δ^{15}N values. Although we did not check the relationship between them in this study, a significantly linear relation was observed for urban and forested mosses in South China when the ratio increased from 1.6 to 6.5 [*Xiao et al.*, 2010a].

[23] The two main sources of NH_y in the atmosphere are those emitted from soils and sewage: the former is less ^{15}N-depleted and the latter more ^{15}N-depleted [*Freyer*, 1978; *Heaton*, 1987]. Our recent studies in Guiyang area showed that moss δ^{15}N value was also a good indicator for different sources of NH_y-N in atmospheric deposition [*Liu et al.*, 2008]. Because moss δ^{15}N values in the

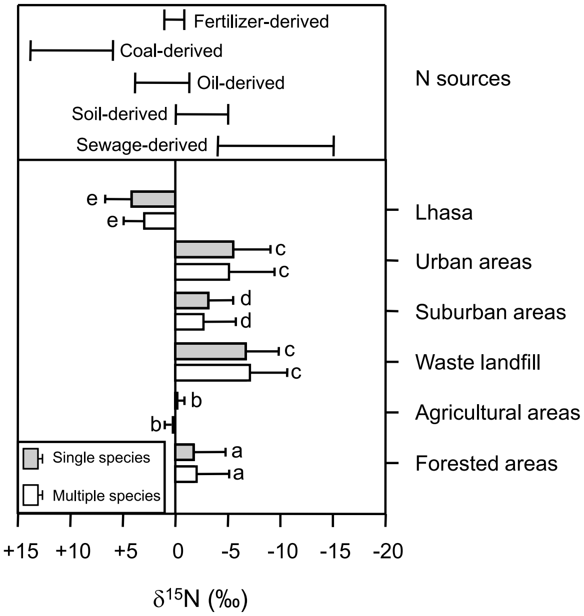

Figure 5. Moss δ^{15}N values in South China. Data from different sampling sites that do not share a letter below the bars are significantly different at $p < 0.05$. The δ^{15}N values of N sources are cited from *Moore* [1977], *Freyer* [1978], *Heaton* [1987, 1990] and *Liu et al.* [2006].

Guiyang area were mainly regulated by NH_y–N from sewage sources (excretory wastes: $-15 \sim -4$‰) and soil sources ($-5.8 \sim -3.3$‰) [*Freyer*, 1978; *Heaton*, 1987], variations of moss δ^{15}N values from the urban to the rural areas were believed to be controlled by the ratios of sewage-derived NH_y to soil-derived NH_y in N deposition. The distinctly different δ^{15}N signatures between urban and forested mosses in the Yangtze River drainage basin indicated that NH_y sources in most of cities and forests were excretory wastes and soil emissions, respectively, because of the similar negative moss δ^{15}N values to those of NH_y sources [*Xiao et al.*, 2010a]. From Figure 5, we could divide the five NH_y-dominated studied areas into four groups based on the moss δ^{15}N values: (1) forested areas (-3‰ to $+3$‰). Mosses in the areas derived less ^{15}N-depleted NH_y emitted from soils; (2) agricultural areas (-1‰ to $+1$‰). Nitrogen in mosses in the areas mainly came from chemical fertilizer; (3) sewage landfill and urban areas (-10‰ to -3‰). More ^{15}N-depleted sewage-derived NH_y was the main N source in these two areas; (4) suburban areas (-5‰ to -1‰). Mosses in the areas were affected by both soil-derived and sewage-derived NH_y.

4.3. Factors Affecting the Estimation of Atmospheric N Deposition

[24] It has to be mentioned that the relationship between N deposition and moss tissue N contents is not necessarily linear (e.g., line J, Figure 3). For instance, *Bragazza et al.* [2005] found that N contents in mixtures of *Sphagnum* mosses decreased logarithmically along the gradient of N deposition. Some factors as discussed below might affect the relationship between N deposition and tissue N contents of mosses.

[25] First, there may be differences between moss species in the accumulation rates of the different forms of nitrogen and in their ability to withstand high levels of N deposition [*Pitcairn et al.*, 2006; *Solga et al.*, 2006]. Therefore, the relationship between N deposition and moss N contents as a guide to N deposition might be species-specific. However, although line C and line B and also line D and line E were based on the same moss species (*Hylocomium splendens* and *Pleurozium schreberi*, respectively), N deposition estimated differed significantly (Figure 3a). It maybe suggested that moss species was not the most important factor when calculating N deposition with the relationship. This has also been proofed by that the correlation coefficients (R^2) of multiple moss species were usually higher than those of single moss species, as summarized in Table 1. Furthermore, atmospheric N deposition estimated using different equations of single moss species (Figure 3a) showed a much wider range than those using different equations of multiple moss species (Figure 3b).

[26] Second, data from livestock studies and the earlier regional study showed that the relationship was not linear at larger inputs because physiological saturation may have occurred at larger N inputs [*Pitcairn et al.*, 1998] and thus the foliar N increases became progressively smaller. However, via *Dicranum polysetum* and *Haplocladium microphyllum*, respectively, *Salemaa et al.* [2008] and *Xiao et al.* [2010a] found that a wider linear range for both N deposition (>50 kg N ha^{-1} yr^{-1}) and moss tissue N contents still existed even when moss N contents reached >3.5%, suggesting that physiological saturation might be only important for some species.

[27] Third, nitrogen uptake in mosses varied according to local factors such as microclimate (precipitation), topography (altitude) [*Zechmeister et al.*, 2008], vegetation, etc. [*Hicks et al.*, 2000; *Pitcairn et al.*, 2006]. But a linear correlation integrated with the data from a wide range of areas was found by *Xiao et al.* [2010a], indicating no importance of local factors when estimating N deposition.

[28] In conclusions, both tissue N contents and ^{15}N natural abundance in epilithic mosses can be accepted as good bio-monitors of atmospheric N deposition in the six kinds of South Chinese areas. The high tissue N contents in South Chinese mosses indicated high N deposition levels in these regions. Assessment of ten previously reported relationships between moss N contents and N deposition suggested that those of multiple moss species were preferred. Mosses in these areas contained isotopically distinct signals, which could distinguish N sources such as sewage-derived NH_y, soil-derived NH_y, chemical fertilizer and NO_x from fuels combustion.

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References

Aber, J. D., W. McDowell, K. Nadelhoffer, A. Magill, G. Berntson, M. Kamakea, S. McNulty, W. Currie, L. Rustad, and I. Fernandez (1998), Nitrogen saturation in temperate forest ecosystems, *BioScience*, *48*, 921–934, doi:10.2307/1313296.

Asman, W. A. H., M. A. Sutton, and J. K. Schjorring (1998), Ammonia: Emission, atmospheric transport and deposition, *New Phytol.*, *139*, 27–48, doi:10.1046/j.1469-8137.1998.00180.x.

Bobbink, R., M. Hornung, and J. G. M. Roelofs (1998), The effects of airborne nitrogen pollutants on species diversity in natural and semi-natural European vegetation, *J. Ecol.*, *86*, 717–738, doi:10.1046/j.1365-2745.1998.8650717.x.

Bragazza, L., et al. (2005), Nitrogen concentration and δ^{15}N signature of *ombrotrophic Sphagnum* mosses at different N deposition levels in Europe, *Global Change Biol.*, *11*, 106–114, doi:10.1111/j.1365-2486.2004.00886.x.

Fan, J. L., Z.-Y. Hu, S.-Y. Zhuang, J. Zhou, T.-J. Wang, and C.-Y. Liu (2007), Observation of atmospheric nitrogen deposition into forestland (in Chinese), *China Environ. Sci.*, *27*(1), 7–9.

Freyer, H. D. (1978), Seasonal trends of NH_4^+ and NO_3^- nitrogen isotope composition in rain collected at Jülich, Germany, *Tellus*, *30*, 83–92, doi:10.1111/j.2153-3490.1978.tb00820.x.

Freyer, H. D. (1991), Seasonal variation of ^{15}N/^{14}N ratios in atmospheric nitrate species, *Tellus, Ser. B*, *43*, 30–44.

Galloway, J. N., W. H. Schlesinger, H. Levy II, A. Michaels, and J. L. Schnoor (1995), Nitrogen fixation: Anthropogenic enhancement-environmental response, *Global Biogeochem. Cycles*, *9*, 235–252, doi:10.1029/95GB00158.

Garten, C. T. (1992), Nitrogen isotope composition of ammonium and nitrate in bulk precipitation and forest throughfall, *Int. J. Anal. Chem.*, *47*, 33–45, doi:10.1080/03067319208027017.

Gerdol, R., L. Bragazza, R. Marchesini, A. Medici, P. Pedrini, S. Benedetti, A. Bovolenta, and S. Coppi (2002), Use of moss (*Tortula muralis* Hedw.) for monitoring organic and inorganic air pollution in urban and rural sites in northern Italy, *Atmos. Environ.*, *36*, 4069–4075, doi:10.1016/S1352-2310(02)00298-4.

Harmens, H., G. Mills, F. Hayes, P. Williams, L. De Temmerman, and The Participants of ICP Vegetation (2005), Air pollution and vegetation: ICP vegetation annual report 2004/2005, report, UNECE ICP Veg. Coord. Centre, Cent. for Ecol. and Hydrol., Bangor, U. K. [Available at http://icpvegetation.ceh.ac.uk.]

Heaton, T. H. E. (1986), Isotopic studies of nitrogen pollution in the hydrosphere and atmosphere: A review, *Chem. Geol.*, *59*, 87–102, doi:10.1016/0168-9622(86)90059-X.

Heaton, T. H. E. (1987), ^{15}N/^{14}N ratios of nitrate and ammonium in rain at Pretoria, South Africa, *Atmos. Environ.*, *21*, 843–852, doi:10.1016/0004-6981(87)90080-1.

Heaton, T. H. E. (1990), ^{15}N/^{14}N ratios of NO_x from vehicle engines and coal-fired power stations, *Tellus, Ser. B*, *42*, 305–307.

Heaton, T. H. E., B. Spiro, S. Madeline, and C. Robertson (1997), Potential canopy influences on the isotopic composition of nitrogen and sulphur in atmospheric deposition, *Oecologia*, *109*, 600–607, doi:10.1007/s004420050122.

Hicks, W. K., I. D. Leith, S. J. Woodin, and D. Fowler (2000), Can the foliar nitrogen concentration of upland vegetation be used for predicting atmospheric nitrogen deposition? Evidence from field surveys, *Environ. Pollut.*, *107*, 367–376, doi:10.1016/S0269-7491(99)00166-9.

Lilleskov, E. A., T. J. Fahey, T. R. Horton, and G. M. Lovett (2002), Belowground ectomycorrhizal fungal community change over a nitrogen deposition gradient in Alaska, *Ecology*, *83*, 104–115, doi:10.1890/0012-9658(2002)083[0104:BEFCCO]2.0.CO;2.

Liu, C.-Q., S.-L. Li, Y.-C. Lang, and H.-Y. Xiao (2006), Using δ^{15}N- and δ^{18}O-values to identify nitrate sources in karst ground water, Guiyang, southwest China, *Environ. Sci. Technol.*, *40*, 6928–6933, doi:10.1021/es0610129.

Liu, X.-Y., H.-Y. Xiao, C.-Q. Liu, and Y.-Y. Li (2007), δ^{13}C and δ^{15}N of moss (*Haplocladium microphyllum* (Hedw.) Broth) for indicating environment variations and canopy retention on atmospheric nitrogen deposition, *Atmos. Environ.*, *41*, 4897–4907, doi:10.1016/j.atmosenv.2007.02.004.

Liu, X.-Y., H.-Y. Xiao, C.-Q. Liu, Y.-Y. Li, and H.-W. Xiao (2008), Stable carbon and nitrogen isotopes of the moss *Haplocladium microphyllum* in an urban and a background area (SW China): The role of environmental conditions and atmospheric nitrogen deposition, *Atmos. Environ.*, *42*, 5413–5423, doi:10.1016/j.atmosenv.2008.02.038.

Moore, H. (1974), Isotopic measurement of atmospheric nitrogen compounds, *Tellus*, *26*, 169–174.

Moore, H. (1977), The isotopic composition of ammonia, nitrogen dioxide, and nitrate in the atmosphere, *Atmos. Environ.*, *11*, 1239–1243, doi:10.1016/0004-6981(77)90102-0.

Pearson, J., D. M. Wells, K. J. Seller, A. Bennett, A. Soares, J. Woodall, and M. J. Ingrouille (2000), Traffic exposure increases natural ^{15}N and heavy metal concentrations in mosses, *New Phytol.*, *147*, 317–326, doi:10.1046/j.1469-8137.2000.00702.x.

Pitcairn, C. E. R., D. Fowler, and J. Grace (1995), Deposition of fixed atmospheric nitrogen and foliar nitrogen content of bryophytes and *Calluna vulgaris* (L.) Hull, *Environ. Pollut.*, *88*, 193–205, doi:10.1016/0269-7491(95)91444-P.

Pitcairn, C. E. R., I. D. Leith, L. J. Sheppard, M. A. Sutton, D. Fowler, R. C. Munro, S. Tang, and D. Wilson (1998), The relationship between nitrogen deposition, species composition and foliar nitrogen concentrations in woodland flora in the vicinity of livestock farms, *Environ. Pollut.*, *102*(1), 41–48, doi:10.1016/S0269-7491(98)80013-4.

Pitcairn, C. E. R., I. D. Leith, M. A. Sutton, D. Fowler, K. J. Hargreaves, M. Moghaddam, V. H. Kennedy, and L. Grannat (2001), Foliage nitrogen as an indicator of nitrogen deposition and critical loads exceedance on a European scale, *Water Air Soil Pollut.*, *130*, 1037–1042, doi:10.1023/A:1013908312369.

Pitcairn, C. E. R., U. M. Skiba, M. A. Sutton, D. Fowler, R. Munro, and V. K. Kennedy (2002), Defining the spatial impacts of poultry farm ammonia emissions on species composition of adjacent woodland groundflora using Ellenberg indicators, nitrous oxide and nitric oxide and foliar nitrogen as marker variables, *Environ. Pollut.*, *119*, 9–21, doi:10.1016/S0269-7491(01)00148-8.

Pitcairn, C. E. R., D. Fowler, I. D. Leith, L. J. Sheppard, M. A. Sutton, V. Kennedy, and E. Okello (2003), Bioindicators of enhanced nitrogen deposition, *Environ. Pollut.*, *126*, 353–361, doi:10.1016/S0269-7491(03)00248-3.

Pitcairn, C., D. Fowler, I. D. Leith, L. J. Sheppard, M. A. Sutton, V. Kennedy, and E. Okello (2006), Diagnostic indicators of elevated nitrogen deposition, *Environ. Pollut.*, *144*, 941–950, doi:10.1016/j.envpol.2006.01.049.

Salemaa, M., R. Mäkipää, and J. Oksanen (2008), Differences in the growth response of three bryophyte species to nitrogen, *Environ. Pollut.*, *152*, 82–91, doi:10.1016/j.envpol.2007.05.019.

Solga, A., J. Burkhardt, H. G. Zechmeister, and J. P. Frahm (2005), Nitrogen content, ^{15}N natural abundance and biomass of the two pleurocarpous mosses *Pleurozium schreberi* (Brid.) Mitt and *Scleropodium purum* (Hedw.) Limpr. in relation to atmospheric nitrogen deposition, *Environ. Pollut.*, *134*, 465–473, doi:10.1016/j.envpol.2004.09.008.

Solga, A., T. Eichert, and J. P. Frahm (2006), Historical alteration in the nitrogen concentration and ^{15}N natural abundance of mosses in Germany: Indication for regionally varying changes in atmospheric nitrogen deposition within the last 140 years, *Atmos. Environ.*, *40*, 8044–8055, doi:10.1016/j.atmosenv.2006.03.024.

Thöni, L., D. Matthaei, E. Seitler, and A. Bergamini (2008), Deposition von Luftschadstoffen in der Schweiz. Moosanalysen 1990–2005, *Umwelt Zustand 0827*, Bundesamt für Umwelt, Bern. [Available at www.umwelt-schweiz.ch/uz-0827-d.]

Vitousek, P. M., J. D. Aber, R. W. Howarth, G. E. Likens, P. A. Matson, D. W. Schindler, W. H. Schlesinger, and D. G. Tilman (1997), Human alteration of the global nitrogen cycle: Sources and consequences, *Ecol. Appl.*, *7*, 737–750.

Xiang, R.-J., L.-Y. Chai, G. Zhang, X.-L. Zhang, and M. Zeng (2006), Input-output dynamics of nitrogen and sulfur in Caijiatang forested catchment in Hunan Province (in Chinese with English abstract), *Acta Sci. Circumstantiae*, *26*(8), 1372–1378.

Xiao, H.-Y., and C.-Q. Liu (2002), Sources of nitrogen and sulfur in wet deposition at Guiyang, Southwest China, *Atmos. Environ.*, *36*, 5121–5130, doi:10.1016/S1352-2310(02)00649-0.

Xiao, H.-Y., and C.-Q. Liu (2004), Chemical characteristics of water-soluble components in TSP over Guiyang, SW China, 2003, *Atmos. Environ.*, *38*, 6297–6306, doi:10.1016/j.atmosenv.2004.08.033.

Xiao, H.-Y., C.-G. Tang, H.-W. Xiao, X.-Y. Liu, and C.-Q. Liu (2010a), Mosses indicating atmospheric nitrogen deposition and sources in the Yangtze River drainage basin, China, *J. Geophys. Res.*, *115*, D14301, doi:10.1029/2009JD012900.

Xiao, H.-Y., C.-G. Tang, H.-W. Xiao, X.-Y. Liu, and C.-Q. Liu (2010b), Stable sulphur and nitrogen isotopes of the moss *Haplocladium microphyllum* at urban, rural and forested sites, *Atmos. Environ.*, *44*, 4312–4317, doi:10.1016/j.atmosenv.2010.05.023.

Zechmeister, H. G., D. Hohenwallner, S. Smidt, I. Roder, S. Maringer, A. Richter, and W. Wanek (2008), Total nitrogen content and δ^{15}N signatures in moss tissue: Indicative value for nitrogen deposition patterns and source allocation on a nation-wide scale, *Environ. Sci. Technol.*, *42*, 8661–8667, doi:10.1021/es801865d.

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