

1 **The fate of ¹⁵N-nitrate in mesocosms from five European**
2 **peatlands differing in long-term nitrogen deposition rate**

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1 **Abstract**

2 Elevated nitrogen (N) deposition changes the retention, transformation, and fluxes of N in
3 ombrotrophic peatlands. To evaluate such effects we applied a ^{15}N tracer ($\text{NH}_4^{15}\text{NO}_3$) at a
4 rate of $2.3 \text{ g N m}^{-2} \text{ yr}^{-1}$ to mesocosms of five European peatlands with differing long-term
5 N deposition rates for a period of 76 days of dry and 90 days of wet conditions. We
6 determined background N content and moss length growth, and recovered the ^{15}N tracer
7 from the mosses, graminoids, shrubs, the peat, and dissolved N. Background N contents
8 in *Sphagnum* mosses increased from 5.5 (Degerö Stormyr, deposition $< 0.2 \text{ g N m}^{-2} \text{ yr}^{-1}$)
9 up to 12.2 mg g^{-1} (Frölichshaier Sattelmoor, $4.7\text{-}6.0 \text{ g N m}^{-2} \text{ yr}^{-1}$). In peat from Degerö
10 nitrate and ammonium concentrations were below 3 mg L^{-1} , whereas up to 30 mg L^{-1}
11 (nitrate) and 11 mg L^{-1} (ammonium) was found in peat from Frölichshaier Sattelmoor.
12 *Sphagnum* mosses (down to 5 cm below surface) generally intercepted large amounts of
13 ^{15}N ($0.2 - 0.35 \text{ mg g}^{-1}$) and retained the tracer most effectively relative to their biomass.
14 Similar quantities of the ^{15}N were recovered from the peat, followed by shrubs,
15 graminoids and the dissolved pool. At the most polluted sites we recovered more ^{15}N
16 from shrubs (up to 12.4%) and from nitrate and ammonium (up to 0.7%). However, no
17 impact of N deposition on ^{15}N retention by *Sphagnum* could be identified and their length
18 growth was highest under high N background deposition. Our experiment suggests that
19 the decline in N retention at levels above ca. $1.5 \text{ g m}^{-2} \text{ yr}^{-1}$, as expressed by elevated near-
20 surface peat N content and increased dissolved N concentrations, is likely more modest
21 than previously thought. This conclusion is related to the finding that *Sphagnum* species
22 can apparently thrive at elevated long-term N deposition rates in European peatlands.

23

24 **1 Introduction**

25 Peatlands, as important carbon (C) stores, have accumulated approximately 615 GtC
26 since the Last Glacial Maximum (Yu et al. 2010). Their carbon uptake can be altered by
27 hydrological, climatic and human-induced factors, such as groundwater level change (e.g.
28 Blodau et al. 2004; Deppe et al. 2010; Moore and Knowles 1989), temperature
29 fluctuations (e.g. Daulat and Clymo 1998; Lafleur et al. 2005) or elevated nitrogen (N)
30 deposition (e.g. Bragazza et al. 2006; Juutinen et al. 2010; Keller et al. 2005). Increase in

1 N concentrations, a key growth-limiting nutrient in peatlands (Aerts et al. 1992), can
2 cause changes in vegetation composition (e.g. Heijmans et al. 2001; Juutinen et al. 2010;
3 Wu et al. 2015) and promote biodiversity loss (Dise et al. 2011).

4 Ombrotrophic peatlands are exclusively fed by rain water and especially sensitive to
5 atmospheric N inputs. Over the last century many of these ecosystems have received
6 inputs of atmospheric N deposition even ten-fold larger than the pre-industrial levels (van
7 Aardenne et al., 2001). In Europe, where N deposition strongly varies spatially, the mean
8 change in N deposition between 1950-2000 and 1900-1950 was between 0.4 and 1.6 g m⁻²
9 yr⁻¹ (Granath et al. 2014). Although the future estimates of N emissions (NO_x and NH₃)
10 vary, with some indicating a substantial further increase (Galloway et al. 2004) and
11 others suggesting stabilization or even slight decrease of emissions by 2030 given an
12 appropriate emission control (Cofala et al. 2007), the importance of elevated N inputs to
13 peatlands remains undisputable.

14 Elevated N supply has been documented to cause various shifts in peatland N cycling
15 such as a decrease of C:N ratio in near surface peat (Bragazza et al. 2012), stimulation of
16 denitrification (Francez et al. 2011), and an increase of N concentration in interstitial
17 waters (Limpens et al. 2003). It has also been reported that plant community change from
18 *Sphagnum*-dominated to vascular-dominated and the enhanced N inputs may turn
19 peatlands into C sources (Bubier et al. 2007; Wiedermann et al. 2007). The effect of N
20 deposition on the retention ability of *Sphagnum* mosses is also dependent on N deposition
21 rate. At low N deposition levels the growth of *Sphagnum* can be promoted (Aerts et al.
22 1992), which suggests that also N retention is raised. At high N deposition, however,
23 *Sphagnum* was found to lose its filtering capacity (Lamers et al. 2000, Limpens and
24 Berendse 2003). As a result elevated N input to ombrotrophic peatlands and declining
25 uptake by *Sphagnum* could potentially enhance denitrification due to increased N
26 availability in the pore waters of affected systems (Hayden and Ross 2005).

27 A quantification of N transformation rates and fluxes under conditions of varying long-
28 term N deposition is crucial for understanding the mechanisms and robustness of N
29 retention. Several experimental studies have addressed the impact of increased N
30 deposition on the functioning of peatland vegetation (e.g. Aldous 2002a; Curtis et al.

1 2005; Williams et al. 1999b). Lower N retention in the upper stem moss tissue (Aldous
2 2002b) and higher N₂O emissions (Regina et al. 1996) have been documented with
3 decreasing water table. However, to our knowledge, the fate of N in peatland ecosystems
4 under varying N inputs and hydrological conditions has not been examined extensively
5 yet (Blodau et al. 2006; Xing et al. 2010). Studies on the fate of N within moss, vascular
6 plants, and upper peat layer, including the pore water, are needed in light of the
7 continued, high levels of N emissions. In this contribution, we investigate how the
8 allocation of N in plants and peat is altered under different N pollution levels using ¹⁵N
9 labeled nitrate. We conducted a controlled mesocosm experiment with intact peat cores,
10 including the vegetation, from five European peatlands, which have a history of differing
11 long-term N deposition. This approach allowed us to compare the fate of deposited ¹⁵N at
12 sites that have undergone decades of differing N deposition and this way to avoid the
13 shortcomings of short-term fertilization studies, in which the time scale of manipulation
14 is generally too short for an adjustment of ecosystem structure and functioning. Labeled
15 N was used to investigate how the allocation of N in plants and peat was altered under the
16 different long-term N pollution levels the sites were exposed to. We hypothesized that the
17 sites with a legacy of low N deposition would experience low mobility and high retention
18 of N in vegetation and near-surface peat, whereas the sites with a legacy of higher N
19 deposition would experience high mobility and low retention of the element. We further
20 expected nearly complete absorption of the applied ¹⁵N by *Sphagnum* at low background
21 N deposition, and diminished absorption by mosses and raised uptake by vascular plants
22 at background N deposition exceeding levels of about 1.0 - 1.5 g N m⁻² yr⁻¹ (Bragazza et
23 al. 2006, Lamers et al. 2000).

24 To address the hypothesis, we specifically examined:

- 25 1. differences in elemental N content in the peat and vegetation before the
26 experiment and dissolved nitrate, ammonium, dissolved organic, and total
27 nitrogen concentrations during the experiment,
- 28 2. differences in the distribution of ¹⁵N stemming from experimentally deposited
29 ¹⁵N-nitrate between plant functional types, peat, and dissolved nitrate and
30 ammonium, and

1 3. differences in the ‘efficiency’ of the vegetation and peat to retain the deposited
2 ¹⁵N during the experiment.

3

4 **2 Materials and Methods**

5 **2.1 Study sites**

6 We selected five *Sphagnum*-dominated peatlands in Northern and Western Europe:
7 Degerö Stormyr (DS), Sweden; Lille Vildmose (LV), Denmark; Fenn's Whixall and
8 Bettisfield Mosses NNR (WM), United Kingdom; Cors Fochno (CF), Wales, United
9 Kingdom; and Frölichshaier Sattelmoor (FS), Germany. The differences in the soil
10 acidity between the sites were small – pH ranged between 3.6 and 4.0 and was typical for
11 ombrotrophic peatlands (Rydin and Jeglum 2006). Site specific characteristics as well as
12 the typical vegetation are presented in Table 1.

13 **2.2 Experimental design and sampling**

14 In the summer of 2009 we collected three intact peat monoliths (“cores”, “mesocosms”,
15 30 cm in diameter, 40 cm long) from lawns of the five sites. This resulted in 15
16 mesocosms, which were subsequently incubated in a greenhouse chamber at 20°C, c.
17 60% rH, and were exposed to 12h light/dark cycles. Directly after collection, all
18 mesocosms were drained and then rewetted to a water table of 28 cm±3 cm below the
19 moss layer. The experiment commenced in November 2009, after a two to three month
20 (depending on the collection date) acclimation period. During the first 76 days all
21 mesocosm were subjected to low water table level (-28 cm±3 cm), here also called ‘dry
22 period’. It is important to note that the moss layer and peat remained moist even during
23 this period due to frequent irrigation. Between day 77 and 78 we raised the water level up
24 to ca. -8 cm using deionised water and maintained it there (±3 cm) for the next 90 days,
25 until the end of the experiment in the middle of April (‘wet period’).

26 Mesocosms were watered twice a week using a self-made sprinkler. During the
27 acclimation period we added a synthetic rainwater solution, whose chemical composition
28 was based on the average European rainwater composition (Appelo and Postma 2005),
29 with N concentrations adjusted for each site. Specifically, the concentrations were 16.65

1 mg L⁻¹ NH₄¹⁵NO₃, and 2.94 (CaCl₂*2H₂O), 1.63 (MgCl), 3.70 (Na₂SO₄), 0.52 (K₂SO₄),
2 0.029 (H₂SO₄) and 0.002 (HCl) mg L⁻¹ during the experiment. In the acclimation period
3 N deposition differed between sites and was adjusted to 1.57 (DS), 6.26 (CF), 9.39
4 (WM), 12.53 (LV) and 16.44 (FS) mg L⁻¹ NH₄NO₃. Throughout the experiment
5 (November-April), we substituted NH₄NO₃ in the irrigate with the NH₄¹⁵NO₃ tracer at 2.3
6 g of N m⁻² yr⁻¹ (1.1 g of N m⁻² added over the experimental period) to each mesocosm.
7 This deposition level was found by Blodau et al. (2006) to be almost fully absorbed by
8 mosses in mesocosms from unpolluted sites, whereas breakthrough of the tracer was
9 expected in mesocosms from N polluted sites. Each tracer application (48 in total) was
10 followed by an addition of distilled water to the cores using the sprinkler. This procedure
11 was adopted to rinse off the tracer from the plant surfaces and to adjust the water table.
12 While this procedure cannot guarantee that no tracer remained on plant surfaces, the large
13 number of irrigation and rinsing events should ensure that only little of the applied ¹⁵N
14 tracer remained physically attached to the plant surfaces by the termination of the
15 experiment and sampling of biomass. The ¹⁵N application thus allowed us to trace the
16 fate of N in peat cores exposed to different background N-deposition levels. The
17 remaining rainwater solution stayed unchanged. When referring to the ‘start’ and the
18 ‘end’ of the experiment we refer to the first ¹⁵N tracer application and the sampling
19 following the last ¹⁵N application, respectively.

20 Throughout the experiment, we recorded the water level in all mesocosms twice a week,
21 and adjusted it in the outlined way. Peat soil moisture (SM) content was logged hourly
22 (ECH₂O EC-5 FD sensors, Em5b loggers, Decagon Devices Inc.) at 4 depths in one core
23 from each site. We calibrated the sensors for each site and for the top and the bottom of
24 the peat core (see Supplementary Material). Polynomial equations ($R^2 > 0.82$) were used
25 to calculate the volumetric water content at 1-hour time steps.

26 Sampling of peat gas and water was performed fortnightly. The extracted gas volume was
27 replaced with nitrogen gas (N₂) at each sampling depth. Gas flux from the mesocosm
28 surface was measured during 20 minutes long sampling period, including 5 sampling
29 points in 5-minute intervals. We used plexiglas chambers (27 cm in height and 29 cm in
30 diameter) with an inlet and outlet tube at the top. These data will be reported in a separate
31 contribution (in preparation). The peat water was sampled using Rhizon® samplers

1 (polymer, 1µm pore size, Eijkelkamp Agrisearch Equipment) at 4 depths: 9 cm, 16 cm,
2 23 cm and 30 cm below the *Sphagnum* surface and through the PE tube from the bottom
3 of each core. The pH of peat water was measured immediately using a glass electrode.
4 The water samples were stored cool, or frozen for later analyses.

5 The length growth of *Sphagnum* moss community was measured in all mesocosms using
6 three cranked wires (Clymo 1970) per mesocosm. Wire lengths were measured shortly
7 after installing, in the middle and in the end of the experiment. Single species of
8 *Sphagnum*, shrubs and graminoids designated for isotopic analyses were sampled from
9 each mesocosm at the end of dry and at the end of wet period. Plant samples used for
10 estimation of background ¹⁵N contents were collected directly from the sampling sites.

11 After termination of the experiment in April 2010 total aboveground biomass of each
12 core was quantified. We clipped the whole plant material from the mesocosms' surface,
13 distinguishing between main plant functional types (shrubs, graminoids and mosses).
14 Mosses were collected down to 5 cm below the moss canopy. After the biomass
15 collection, we sequentially extracted the pore water from individual depth layers through
16 the gas sampler openings. The extraction continued from the top of the core downwards
17 until no water from the single layer could be obtained. Due to a large water volume
18 required for the ¹⁵N analysis and differences in a field capacity between the cores and
19 specific peat layers, it was not possible to obtain exactly the same layer depths for all
20 mesocosms. At last, the cores were extracted and one part of each core cut into 2 - 8 cm
21 (higher resolution at the top and lower at the bottom) segments for ¹⁵N analyses. The
22 remaining parts were used for bulk density (BD) estimations. The peat for background
23 ¹⁵N measurements was collected before the first tracer application using a stainless steel
24 microcorer (3 cm in diameter).

25 All organic samples were dried at 70°C for at least 24 hours or until no more mass loss
26 could be recorded. The samples used for aboveground biomass and bulk density
27 estimation were then weighed and the samples destined for isotopic analysis were ground
28 with a ball mill to fine powder and stored in an evacuated desiccator prior to analysis.

1 When reporting sampling depths, we report the depth relevant not to the rim of the
2 mesocosm tube but to the *Sphagnum* surface, thus obtaining discrepancies of up to 1 cm
3 between the cores.

4 **2.3 Chemical analyses**

5 In the peat pore water obtained by rhizon samplers we measured nitrate (NO_3^-)
6 concentrations by ion chromatography (Metrohm IC 733 with suppressor module,
7 Metrosep Dual 3 column). Ammonium (NH_4^+) concentrations were quantified
8 photometrically at 690 nm (ISO 7150/1) on a DR 3800 photometer using NH_4^+ quick test
9 method (Ammonium test, range: 0.01-3.00 $\text{NH}_4\text{-N mg L}^{-1}$, Merck). Total dissolved N
10 (TDN) and particulate N (PN) were measured on a TOC/TN analyzer (multi N/C 2100,
11 Analytik Jena, NPOC analysis). Dissolved organic N (DON) concentrations were
12 calculated as the difference between TDN and dissolved inorganic N (DIN) (NO_3^- plus
13 NH_4^+). In peat pore gases obtained by silicon gas samplers and in chamber measurements
14 nitrous oxide (N_2O) was measured on a Varian gas chromatograph with flame ionization
15 (FID) and electron capture (ECD) detectors. We did not detect any N_2O above
16 background in the pore water and flux from the mesocosms throughout the experiment;
17 data on N_2O are thus not reported in this contribution.

18 **2.4 Isotopic analyses**

19 The ratios of natural abundance and tracer $^{15}\text{N}/^{14}\text{N}$ as well as total C and N were
20 quantified in the dried plant material and the peat using an elemental analyzer (NA 1108
21 – natural samples, NC 2500-labeled samples, CE instruments, Milano, Italy), connected
22 via ConFlo III interface to a delta S/delta plus IRMS (natural/labeled, Finnigan/Thermo
23 Fisher Scientific, Bremen, Germany). Dissolved inorganic ^{15}N content in pore water
24 samples was assessed by a sequential trapping of volatilized NH_4^+ and NO_3^- (Brooks et
25 al. 1989) - procedure modified and described in detail in Blodau et al. (2006).

26 High purity of the added tracer (98 atom% ^{15}N) and high frequency of watering events
27 resulted in extremely high isotopic signatures of most peat and plant samples, exceeding
28 the calibration range of the instrument. We thus diluted all samples with a natural plant
29 material of known ^{15}N content and re-measured the samples. Comparison of the two data

1 sets revealed a reasonably close correlation ($y = 0.9623x + 0.0036$, $R^2 = 0.71$, see
2 Supplementary Material, Fig. S1). For interpretation we thus used the original ^{15}N data
3 set.

4 The ^{15}N results were expressed as % content of ^{15}N atoms in total N atoms (atom% $^{15}\text{N} =$
5 $[\text{}^{15}\text{N}/(\text{}^{14}\text{N}+\text{}^{15}\text{N})] \times 100$ atom%). The ^{15}N abundances (atom% ^{15}N excess) were obtained
6 by subtracting the background ^{15}N of plants, peat and traps (diffusion procedure) from the
7 labeled samples.

8 **2.5 Data handling and statistical analyses**

9 We calculated area-based plant biomass, peat density and total nitrogen (TN) content of
10 the ecosystem pools as well as volume-based ^{15}N content in peat and plant pools.
11 Additionally, ^{15}N retention efficiency and ^{15}N recovery from the pools was estimated.
12 The relative share of each of the ecosystem pools to the total ^{15}N recovered (%) was
13 obtained by dividing their tracer mass by the total mass of the tracer added to the system.
14 The sum of the relative shares of each pool gave the total ^{15}N recovery.

15 Statistical analyses were performed with the SPSS 20.0 package and SigmaPlot for
16 Windows, version 11.0. The effect induced by differing long-term N deposition (sites) on
17 N pools and ^{15}N retention and of wet and dry conditions on ^{15}N uptake by plants was
18 analyzed by one-way analysis of variance (ANOVA) followed by a post-hoc test (Tukey
19 or Bonferroni). The effect of differing long-term N deposition (sites) on ^{15}N abundance in
20 vegetation pools after the dry and wet phase was analyzed with one-way analysis of
21 covariance (ANCOVA) using wet and dry conditions as covariate and log-transforming
22 data when needed. Significance was set at $p \leq 0.05$, unless stated otherwise.

23 Before reporting the results of N concentration measurements, a few words on the
24 uncertainty in data are needed. We encountered an issue when evaluating the pore water
25 concentration data from FS cores – NO_3^- concentrations in the first part of the experiment
26 by far exceeded the measured TDN concentrations. A repeated measurement of the
27 problematic samples did not give a solution to that problem. Thus, the DON data for the
28 first 98 days needed to be omitted, otherwise showing negative values.

29

1 **3 Results**

2 **3.1 Plant biomass and growth**

3 Total plant biomass at the end of the experiment was largest in the Welsh cores with 1530 ± 313 g
4 m^{-2} (CF) and 1397 ± 693 g m^{-2} (WM) and smallest with 1178 ± 67 g m^{-2} in German Frölichshaier
5 Sattelmoor (FS) mesocosms (Fig. 1a). Bryophytes accounted for 62% to 87% of the total (Fig.
6 1b), when defining living biomass as reaching down to a depth of 5 cm below the *Sphagnum*
7 capitulum. Shrub biomass was significantly higher than graminoid biomass with the exception of
8 Degerö Stormyr (DS) (Fig. 1b). The highest percentage of shrubs (30%) was found in FS cores.
9 Vascular plant biomass increased at all sites during the experiment, particularly after raising the
10 water table. The highest increase – from 140 g DW m^{-2} to 440 g DW m^{-2} – was recorded in FS
11 cores (Supplementary Material, Fig. S2); overall biomass gain decreased in the order
12 FS>WM>CF>LV \cong DS. *Sphagnum* growth differed substantially among the sites. In Frölichshaier
13 Sattelmoor (FS) mean growth rate was 2.36 mm month^{-1} and at Whixall Moss (WM) only 0.27
14 mm month^{-1} ; growth declined in the order FS>DS>LV>CF>WM (Supplementary Material, Fig.
15 S3). During the dry period *Sphagnum* mosses did not desiccate and actively grew, likely due to
16 the frequent irrigation. We measured an increase in growth at CF, WM and Lille Vildmose (LV)
17 cores during wet period (Supplementary Material, Fig. S3), but only for CF this increase was
18 significant ($P < 0.02$).

19 **3.2 Nitrogen contents**

20 Weight based (Fig. 2) and area based (Fig. 3) N contents differed between the sites at the
21 end of the experiment, albeit often not significantly. The average N content of peat, plant
22 and dissolved pools, expressed as the ratio of the mass of N present in the pool to the
23 total mass of the pool, generally increased with long-term N deposition (Fig. 2). Surface
24 peat layer (5-15 cm depth) contained between 5.3 ± 0.3 (DS) and 13.4 ± 2.3 (CF) mg N g^{-1}
25 and this difference was significant. *Sphagnum* (capitulum and stem) and shrubs had
26 similar N contents varying between 4.9 ± 2.5 and 9.8 ± 3.9 mg g^{-1} , and 5.7 ± 0.8 and 10.7
27 ± 1.7 mg g^{-1} respectively. The N content in *Sphagnum* was generally higher in the apical
28 part than in the stem part and ranged between 5.6 ± 0.5 (DS) and 12.2 ± 6.4 (FS) mg N g^{-1}
29 (Supplementary Material, Fig. S5). We recorded a very high shrub N content at Degerö
30 Stormyr resulting from high N content of *Andromeda polifolia* leaves at this site (Fig.2).
31 Graminoids were the most N-rich plant functional type (8.4 ± 1.5 - 18.9 ± 4.0 mg g^{-1}) and

1 their N content differed significantly ($F(4,10) = 8.099, p = 0.004$) between Frölichshaier
2 Sattelmoor and the rest of the sites. The least N was found in the dissolved pool (Fig. 2).
3 Total dissolved N content in the surface layer of peat (0-15 cm) ranged from 1.95 ± 0.12
4 to $6.17 \pm 1.27 \mu\text{g g}^{-1}$ and differed significantly between LV cores and DS and WM cores
5 ($F(4,10) = 5.82, p = 0.01$). ANOVA further showed that N content in the ecosystem pools
6 significantly differed from each other; the post-hoc test indicated that at the level of sites
7 the differences between graminoids, *sphagnum* and shrubs were significant at the DS site
8 ($p = 0.002$ and $p = 0.018$) and FS site ($p = 0.01$ and $p = 0.001$). Significant differences
9 also occurred between graminoids and peat (DS, FS, CF), and shrubs and *sphagnum* and
10 peat (CF).

11 Area based N content of ecosystem pools, calculated by multiplying biomass (plants),
12 bulk density (peat) or volumetric water content (Supplementary Material, Table S1) with
13 N content after termination of the experiment, varied between the sites but the differences
14 were mostly not significant (Fig. 3). Due to its large mass, peat contained by far the
15 largest N pool and held between 22.8 ± 4.6 (DS) and 63.5 ± 11.5 (CF) g N m^{-2} in the 10
16 centimeter subsurface layer. *Sphagnum* moss constituted the second largest N pool and
17 contained between 4.0 ± 0.8 (WM) and 9.0 ± 0.3 (LV) g N m^{-2} . Total N content was
18 similar for shrubs and graminoids varying between 0.6 ± 0.16 and $2.8 \pm 0.61 \text{ g N m}^{-2}$ and
19 between 0.4 ± 0.25 and $1.7 \pm 2.07 \text{ g N m}^{-2}$, respectively. The N content in these two plant
20 functional types was highest at Frölichshaier Sattelmoor (Fig. 3), and statistically differed
21 from that of the DS site. Lowest TDN values were detected at low long-term N
22 deposition for shrubs and at intermediate long-term N deposition levels for graminoids
23 (Fig. 3). The dissolved pool contained the smallest quantities of N. Total dissolved N
24 content increased with long-term N deposition and ranged from 0.03 to 0.63 g N m^{-2} (Fig.
25 3). The largest amounts of TDN were found in the LV site and differed significantly from
26 both DS and WM (Fig. 3).

27 In *Sphagnum* mosses, molar ratios of C/N were significantly related to long-term N deposition
28 at the sites and responded to the experimental treatment with the deposition of the $\text{NH}_4^{15}\text{NO}_3$
29 tracer. Ratios of C/N in *Sphagnum* capitula sampled at the sites before the experiment declined
30 from above 100 (Degerö Stormyr, DS) to 36 in German Frölichshaier Sattelmoor (FS) following
31 a log-function (Supplementary Material, Fig. S4, $R^2 = 0.96, p = 0.00022$). Experimental

1 deposition of $2.3 \text{ g N m}^{-2} \text{ yr}^{-1}$, i.e. 1.1 g N m^{-2} during the experiment, led to a decline of C/N in
2 DS and an increase in FS, and little change otherwise; wet conditions partly reversed this effect
3 (Fig. S4).

4 **3.3 Nitrogen uptake by plants**

5 Uptake of ^{15}N during the experiment varied between plant functional types and was
6 influenced by water table position. The highest ^{15}N enrichment was observed in the
7 *Sphagnum*'s apical part, followed by *Sphagnum* stem, shrubs and graminoids (Fig. 4). N
8 deposition had a significant effect on ^{15}N content in *Sphagnum* capitula (ANCOVA, $p =$
9 0.029) and graminoids ($p = 0.042$) but not on *Sphagnum* stems and shrubs. Enrichment
10 with ^{15}N in *Sphagnum* (0 - 5 cm) also visually decreased with long-term N deposition,
11 with Degerö Stormyr having highest and Frölichshaier Sattelmoo smallest uptake during
12 the dry period at 5.9 and 2.9% in the capitulum, respectively (Fig. 4A). Tracer uptake by
13 the capitulum of *Sphagnum* was significantly elevated during the dry period (Fig. 4A), as
14 confirmed by one-way ANCOVA ($p < 0.001$). Unlike in *Sphagnum*, the largest ^{15}N
15 enrichment in shrubs occurred during the wet period (Fig. 4C and 4D). Shrubs
16 assimilated on average twice as much of the tracer during this second part of the
17 experiment (Fig. 4C; one-way ANCOVA, $p < 0.001$). Effects were not significant in
18 regard to graminoids and *Sphagnum* stems.

19 **3.4 Retention and recovery of added ^{15}N**

20 To examine the retention capabilities of each of the pools during the experiment,
21 retention efficiency per gram of dry weight was calculated from the quotient of ^{15}N
22 retained at the end of the experiment divided by dry biomass or dry peat mass,
23 respectively. The sites substantially differed in ^{15}N retention efficiency, when all pools
24 were accounted for, yet differences between sites were statistically significant only with
25 regard to the shrub pool and sites DS and CF, and DS and WM (Fig. 5). Vegetation was
26 up to four times more effective in retaining N than peat and *Sphagnum* moss was
27 significantly more efficient than graminoids and peat ($p = 0.007$ and $p = 0.001$), as were
28 the shrubs ($p = 0.013$). Cors Fochno and Whixall Moss vegetation had the smallest
29 retention efficiency varying between 0.05 ± 0.02 in graminoids (CF) and 0.2 ± 0.06 in

1 *Sphagnum* (0 - 5 cm) (WM) (Fig. 5). Interestingly, *Sphagnum* from both the sites with
2 low (DS) and the sites with high (LV, FS) level of background N input was equally
3 effective in absorbing N, but peat in LV and FS cores retained substantially less N than
4 the peat in WM cores (Fig. 5).
5 During the course of the experiment we applied 38 mg (572 mg m⁻²) of ¹⁵N to each
6 mesocosm. After 24 weeks, the recovery, i.e. the mass of ¹⁵N divided by amount of ¹⁵N
7 added, of the tracer from all N pools ranged from 87.5 to 131% (Table 2). Recoveries
8 above 100% were most likely caused by the uncertainty in the peat bulk density. The
9 depth resolution used in ¹⁵N sampling (6-8 sections per core) was much higher than the
10 resolution used in the bulk density measurements, which encompassed only 2-3 depth
11 intervals per core. Especially the ¹⁵N content in the most upper peat layers, where BD
12 was possibly lower than the average BD of a thicker peat layer, may have been
13 overestimated. *Sphagnum* and peat contained the largest fraction of the tracer and
14 accounted for 21.9 to 68.9% and 23.6 to 65.6% of the total, respectively. Vascular plants
15 contained only between 5.6 and 13.6% of the recovered ¹⁵N. The shrubs had an up to
16 thirteen times higher share than the graminoids (Table 2). The shrubs in cores from
17 Frölichshaier Sattelmoor were particularly efficient in retaining the tracer and accounted
18 for 91% of the retention in vascular plants (Table 2). Dissolved inorganic ¹⁵N contributed
19 little to the retention of ¹⁵N at levels below 1% but its share increased with background N
20 deposition rate (Table 2). This pool accounted for 0.01% (DS), followed by 0.2% (CF
21 and WM), 0.4% (FS) and 0.7% (LV).

22 **3.5 Dissolved nitrogen**

23 The dissolved N concentration increased with long-term N deposition at the sites during
24 the experiment (Fig. 6, Table 1). Nitrate was detectable predominantly above the water
25 level but remained low (< 3 mg L⁻¹) in the majority of the cores (Fig. 6). Only in the
26 cores from Frölichshaier Sattelmoor, NO₃⁻ concentrations reached 30 mg L⁻¹ and were
27 especially high during the first 35 days of the dry period. Ammonium concentrations
28 reached 11 mg L⁻¹ at the site characterized by high levels of N input (FS) and 2 mg L⁻¹ at
29 the site with low level of N input (DS) (Fig. 6). The smallest detectable NH₄⁺
30 concentrations were found at DS and were as low as 0.3 mg L⁻¹ at a depth of 30 cm half

1 way through the experiment (Fig. 6). The highest NH_4^+ concentrations were found at FS,
2 15 - 20 cm below the *Sphagnum* surface, i.e. ca. 10 - 15 cm above water level, during the
3 dry period (Fig. 6). Between days 77 and 78 ammonium concentrations declined, which
4 was probably caused by a dilution from wetting, but returned to previous levels within 5
5 days (Fig. 6). Total dissolved N remained low at the low long-term N deposition sites
6 (DS) and increased substantially in the high long-term N deposition site (FS)
7 (Supplementary Material, Fig. S6 A). Higher concentrations were found mainly above the
8 water table (Fig. 6). The concentrations varied from 1.3 to 5.1 mg L^{-1} at Degerö Stormyr
9 and from 2.4 to 17.3 mg L^{-1} at Frölichshaier Sattelmoor. Dissolved organic N also
10 increased with long-term N deposition during the experiment (Fig. 6, Table 1). Sites with
11 a legacy of high long-term N deposition (LV, FS) had elevated DON concentrations
12 reaching 5.3 mg L^{-1} (FS), whereas the other sites contained less than 1 mg DON L^{-1} .
13 Particulate N content at the bottom outlet of the cores was between 0.2 and 5 mg L^{-1}
14 (Supplementary Material, Fig. S6 B). The highest PN concentrations, recorded for sites
15 with intermediate and high long-term N deposition levels (WM, LV, FS), were measured
16 during first 15 days of the experiment and declined substantially thereafter
17 (Supplementary Material, Fig. S6 B). At Degerö Stormyr, PN and TDN contents in the
18 pore water were similar and generally did not exceed 3 mg L^{-1} (Supplementary Material,
19 Fig. S6). At other sites, PN was on average 20 to 90% lower than TDN.

20 **3.6 ^{15}N distribution in peat and pore water**

21 The distribution of the experimentally applied ^{15}N in peat was influenced by background
22 N deposition rates and decreased with depth. A sharp decrease of the ^{15}N signal,
23 expressed as the difference between the ^{15}N signal recorded at the end of the experiment
24 and the natural ^{15}N background, was observed between 10 and 15 cm depth and 78 - 98%
25 of the total ^{15}N peat pool was retained in the top 20 cm of the peat profile (Fig. 7). The
26 isotopic signal was elevated most at sites with the low and intermediate long-term N
27 deposition levels and reached 4 at% ^{15}N excess in the near-surface peat (Fig. 7A) at the
28 end of the experiment. At sites with high long-term N deposition rates, on the contrary,
29 the amount of ^{15}N in excess of the background was much lower – in a range of 0.2 - 0.6
30 at% in the top layers (Fig. 7, DS). The mean values oscillated between 0.28 (LV) and

1 1.95 (DS) at% ^{15}N excess. Volume based ^{15}N content in peat was calculated by
2 multiplying the experimental isotopic signal of the ^{15}N content in peat layers by their bulk
3 density, and the pattern was similar to the distribution of the isotopic signal (Fig. 7B).
4 The isotopic signal in pore water was coupled to the ^{15}N distribution in the peat, as
5 contents decreased both with depth and background N deposition (Fig. 8A and 8B). The
6 $^{15}\text{N}\text{-NO}_3^-$ and $^{15}\text{N}\text{-NH}_4^+$ concentrations were only in FS cores lower than expected, which
7 was most probably a result of dilution with abundant, unlabelled dissolved N at this site.
8 The ^{15}N excess in nitrate was similar among sites (Fig. 8A), but it substantially differed
9 regarding ammonium (Fig. 8B). Volume-based dissolved inorganic ^{15}N content – the
10 product of DIN concentration, volumetric water content and at% $^{15}\text{N}\text{-DIN}$ excess –
11 differed significantly (Kruskal-Wallis, $p < 0.001$) between nitrate and ammonium but not
12 between sites (Fig. 8C and 8D). The differences mostly resulted from the discrepancies in
13 concentrations between these two N species (Fig. 6). The ammonium concentration was
14 up to 100 times higher than the corresponding nitrate concentration. Concentration of
15 $^{15}\text{N}\text{-NH}_4^+$ generally decreased with depth, whereas $^{15}\text{N}\text{-NO}_3^-$ did not show any specific
16 pattern and remained at low levels at Degerö Stormyr, Cors Fochno and Whixall Moss,
17 with exception of the bottom layer (Fig. 8C and 8D).
18 A more detailed examination of the individual cores from Degerö Stormyr (low
19 background N deposition) and Frölichshaier Sattelmoor (high background N deposition)
20 revealed two patterns of the dissolved $^{15}\text{N}\text{-NH}_4^+$ distribution in the peat (Fig. 9).
21 Frölichshaier Sattelmoor had up to tenfold larger at% ^{15}N excess than Degerö Stormyr
22 (0.036 – 0.349 at% $^{15}\text{N}\text{-NH}_4^+$ excess and 0.002 – 0.045 at% $^{15}\text{N}\text{-NH}_4^+$ excess
23 respectively). The $^{15}\text{N}\text{-NH}_4^+$ concentration decreased gradually with depth in the FS
24 cores, but peaked between 16 cm and 21 cm below the surface in the DS replicates (Fig.
25 9).

26

27 **4 Discussion**

28 **4.1 Nitrogen transfer within the mesocosm system**

29 The aim of this mesocosm study was to characterize changes in the transport and
30 transformation of N within ombrotrophic peatlands that are induced by long-term N
31 deposition. We are aware of a number of experimental limitations. First of all we did not

1 address all aspects of N pools and transformations – uptake of N by microorganisms and
2 transfer of ^{15}N into DON were, for instance, not investigated. It should be kept in mind as
3 well that the results were obtained with experimental deposition of nitrate, which is
4 generally the dominant inorganic N source in non-agricultural areas (Galloway et al.
5 2004), yet may not be the preferred form of N assimilated by mosses (Wiedermann et al.
6 2009). We also have no means to quantify how much of the applied ^{15}N nitrate remained
7 physically attached on plant surfaces rather than being absorbed, although we attempted
8 to minimize this problem by frequent irrigation and rinsing with distilled water. Despite
9 these limitations, the results at hand provide some insight into the fate of N entering
10 ombrotrophic bogs in the form of nitrate subsequent to varying long-term N deposition
11 rate. While in all cores ^{15}N retention in Sphagnum and peat dominated (Table 2), the
12 distribution of the ^{15}N tracer between the mesocosm pools differed depending on the
13 background N deposition. We found a decline of N recovery from the peat pool and an
14 increase of N recovery from shrubs and the dissolved pool with higher long-term N
15 deposition (Table 2, Table 1). A similar change in the N recovery of added ^{15}N -nitrate
16 was observed by Xing and others (2010) after 7 years in a nutrient fertilization study at
17 the Mer Bleue bog in Ontario, where controls and treatments were fertilized with 1.6, 3.2
18 and 6.4 g N m⁻² yr⁻¹ applied in 7 doses during the vegetation period. The retention
19 efficiency of vegetation pools was lowest in the mesocosms from the British sites
20 characterized by the intermediate long-term N deposition levels (Fig. 5), which is in
21 conflict with expectations. Curtis and others (2005), for example, argued for a retention
22 efficiency peaking at intermediate deposition levels and decreasing towards polluted
23 sites. The data obtained by us do not allow clarifying the reason for this phenomenon.
24 The differences, which were not significant, may just be a result of differences in the
25 biomass pool sizes and differing plant species intercepting the tracer.
26 The peat not only contained the largest N pool (Fig. 3) but also similar or even more ^{15}N
27 than the moss layer (Table 2), which here was defined to reach down to a depth of 5 cm
28 beneath the moss canopy. This finding is in agreement with a previous mesocosm study
29 performed by Blodau et al. (2006) using peat cores and ^{15}N -nitrate as tracer from the Mer
30 Bleue Bog exposed to approx. 1.6 g N m⁻² yr⁻¹ and the Experimental Lake Area (Ontario)
31 exposed to approx. 0.3 g N m⁻² yr⁻¹, and the previously mentioned field study by Xing et

1 al. (2010). A substantial mass transfer of ^{15}N into the peat occurred, although only very
2 little dissolved ^{15}N was recovered (Table 2). Blodau and others (2006) could not explain
3 the N transfer into the peat by solute movement and speculated that ^{15}N was transported
4 in form of the particulate organic N (PON), which was not analysed in their study. The
5 movement of PON could, to some extent, explain high recoveries of ^{15}N from the peat
6 pool also in this case, although no experimental evidence can be provided. One can
7 speculate that the PN concentrations measured at the bottom outlet of the cores were
8 probably only a fraction of those in the upper layers considering that the highest TDN
9 concentrations were measured between 15 cm and 25 cm below the *Sphagnum* surface
10 (Fig. 6).

11 Mosses not only intercepted large amounts of ^{15}N in the mesocosms but they also
12 retained the tracer most effectively relative to their biomass. Since *Sphagnum* serves as
13 an effective filter for inorganic N under unpolluted conditions (Lee et al. 1990), we
14 expected ^{15}N -nitrate to accumulate most strongly in the mosses of Degerö Stormyr (Table
15 1, Fig. 2), and inorganic N concentration to be very low beneath. The Swedish site was
16 indeed most efficient in retaining N, storing on average 0.25 mg N g^{-1} , mostly in
17 *Sphagnum* (0 - 5 cm) and shrubs (Fig. 5). We also recovered 65% of tracer from the
18 Degerö Stormyr *Sphagnum* pool only (Table 2) and measured the highest ^{15}N content in
19 the near-surface peat (Fig. 7) at that site. In line with our hypothesis that N mobility
20 would increase with a legacy of long-term N deposition, particularly above previously
21 found N threshold values of $1.0 - 1.5 \text{ g N m}^{-2} \text{ yr}^{-1}$ (Bragazza et al. 2006, Lamers et al.
22 2000), we found polluted sites (LV and FS) to contain the largest dissolved N pools (Fig.
23 3) and the highest nitrate concentrations (Fig. 6). Nitrate not assimilated by N-rich
24 *Sphagnum* (Fig. 2) leached through the *Sphagnum* layer and entered the rooting zone of
25 the vascular plants. This finding was also reflected in the higher ^{15}N recovery from
26 shrubs with increasing background N deposition (Table 2).

27 Remarkably, however, large amounts of ^{15}N were still retained in the N-rich *Sphagnum*
28 layers. Recoveries of 58% and 45.5% for Lille Vildmose and Frölichshaier Sattelmoor
29 respectively, were in the range of the recovery recorded for the *Sphagnum* layer (0 - 5
30 cm) from Degerö Stormyr. The finding suggests that under N-rich conditions *Sphagnum*
31 mosses can still filter inorganic N, most likely by the ongoing formation of new biomass,

1 which continuously requires N. Recovery of ^{15}N from the *Sphagnum* layer is further in
2 agreement with the *Sphagnum* biomass and growth data obtained in this study. In
3 mesocosms from Lille Vildmose and Frölichshaier Sattelmoor, *Sphagnum* mosses had a
4 similar biomass than in the other mesocosms at the end of the experiment (Fig. 1).
5 Particularly in mesocosms from N-rich Frölichshaier Sattelmoor, moss length growth was
6 strong during the dry and wet period as well (Supplementary Information, Fig. S3). The
7 ongoing ^{15}N uptake by mosses in the experiment thus appears plausible. Our results
8 corroborate previous studies, where high N deposition rates did not seem to have a
9 detrimental effect on *Sphagnum* growth (e.g. Granath et al. 2009, Heijmans et al. 2001,
10 Limpens and Berendse 2003) and other, indirect changes, such as in the abundance of
11 vascular plants, seemed to be more important for *Sphagnum* production (Juutinen et al.,
12 2010, Limpens et al. 2011). When the plant-peat soil system remains intact and
13 dominated by mosses and shrubs, as was the case across the selected sites, a threshold at
14 which the filter function of *Sphagnum* is entirely lost does not seem plausible.
15 The robustness of N uptake by mosses may further be assisted by physiological
16 acclimation and species replacement. We allude that an adjustment to higher N inputs in
17 Lille Vildmose and Frölichshaier Sattelmoor may have contributed to the greater
18 resilience of *Sphagnum* species against enhanced long-term N deposition. The question
19 arises whether a) high N retention in the N-rich *Sphagnum* layers was a consequence of
20 the ability to better utilize N by fast-growing species (*papillosum* (FS), Table 1) or
21 whether b) a shift to more tolerant species had occurred. Heijmans and others (2001)
22 found that *Sphagnum* growth during three growing seasons was not limited by high (5 g
23 $\text{N m}^{-2} \text{ yr}^{-1}$) levels of N deposition and mosses previously exposed to a background
24 deposition of ca. $1.5 \text{ g N m}^{-2} \text{ yr}^{-1}$ were still able to assimilate a large portion of
25 experimentally deposited N. The potential ecophysiological adjustment of mosses to an
26 elevated N deposition could evolve, for example, by a gradual increase of the *Sphagnum*
27 tissue N content (Wiedermann et al. 2009). Given that the species in the Lille Vildmose
28 and Frölichshaier Sattelmoor mesocosms differed, it cannot be ruled out a priori either
29 that a shift to different species assisted in maintaining the filtration capacity of *Sphagnum*
30 at these sites. Historical information of Loeske (1903) does not support this idea,
31 however, because in the beginning of the 20th century *S. papillosum* already dominated in

1 the area of Torfhaus, where the Frölichshaier Sattelmoor mesocosms were sampled.
2 Thus, the scenario of species' shift does not seem credible, at least with regard to the
3 Frölichshaier Sattelmoor peatland.

4 Nitrogen uptake by *Sphagnum* and higher plants was influenced by peat water content.
5 Since there is no mechanism for controlling N uptake in *Sphagnum* (Jauhiainen et al.
6 1998), a prolonged exposure to high ambient N concentrations may threaten their vitality.
7 *Sphagnum* mosses can acquire N in three ways: (1) from airborne drift via wet and dry
8 deposition (e.g. Li and Vitt 1997), (2) by recycling from older parts to the youngest, most
9 actively growing ones (Aldous 2002a; Malmer 1988; Urban and Eisenreich 1988), (3) by
10 N₂ fixation (Damman 1978; Hemond 1983). During the first part of the experiment,
11 under dry conditions but with moss growth ongoing, tracer input from the top was the
12 main N source for mosses. Non-symbiotic N₂ fixation might have also played a role but
13 was not assessed. Evidence for the significance of this process is limited to date.
14 Waughman and Bellamy (1980) found bogs to generally have a low activity of
15 nitrogenase and demonstrated an annual N₂ fixation of 0.07 g N m⁻², which is low in
16 comparison to input by precipitation. However recently, Larmola and others (2013)
17 estimated the annual N₂ fixation at the Finnish peatland to range from 0.1 g N m⁻² to 2.9 g
18 N m⁻² – values up to 10 times greater than the inorganic N deposition rate recorded at the
19 studied region. As for the differing effect of raised water table on ¹⁵N uptake by mosses
20 and vascular plants we can only speculate. More N, particularly ammonium, from deeper,
21 now inundated layers likely reached the living tissue of *Sphagnum* plants. This additional,
22 ¹⁵N-poor influx probably diluted the ¹⁵N input from irrigation resulting in a lower
23 *Sphagnum* ¹⁵N content during the wet period (Fig. 4). Vascular plants, on the contrary,
24 assimilated substantially larger amounts of the tracer during the second part of the
25 experiment (Fig. 4). As the direct absorption of atmospheric N through leaf tissue is
26 minimal, root uptake from pore water is the most common N supply for shrubs (Li and
27 Vitt 1997). Shrubs can experience oxygen deficiency when faced with continuously high
28 water levels since they lack aerenchyma. It seems plausible that inundation led to a
29 relocation of root activity to the upper, aerated peat layers, where more ¹⁵N was present
30 according to our measurements (Figs. 7 and 8). Murphy et al. (2009) suggested that
31 shallow water tables in bog systems promote a shallower root distribution. It is also

1 possible that more of the added ^{15}N surpassed the moss layer and entered the rooting zone
2 of the vascular plants due to a generally raised N supply to the mosses when water table
3 was high.

4 **4.2 Nitrogen transformations and transport in the dissolved phase**

5 The ^{15}N tracer penetrated deeper into the peat and more ^{15}N was recovered from shrubs
6 and graminoids in the cores from the sites with a legacy of strongly elevated N deposition
7 (LV, FS). This finding agrees with the hypothesis of increased N mobility and decreased
8 retention in mosses and near-surface peat with long-term N deposition (Table 1). We did
9 not detect any N_2O fluxes but we observed dissolved inorganic N in the pore water and
10 recorded higher concentrations of organic N at polluted sites. Concentrations of N_2O
11 fluxes are difficult to quantify due to the episodic nature of N_2O production. Moreover,
12 acidic conditions typical for ombrotrophic peatlands can slow down or even inhibit
13 denitrification (Williams et al. 1999a). The increased concentrations of DIN and TDN
14 detected below the *Sphagnum* layer might be an evidence of reduced retention of the
15 *Sphagnum* layer caused by long-term elevated atmospheric N inputs at the study sites
16 (FS). Such an observation was, for example, reported in the studies by Lamers et al.
17 (2000) and Limpens et al. (2003). The concentrations of DON also increased with long-
18 term N deposition (Table 1) and were highest at the Frölichshaier Sattelmoor site (Fig. 6).
19 A similar pattern was recorded by Bragazza and Limpens (2004) during a field study of
20 six European bogs spanning a range of chronic atmospheric N inputs from 0.2 to 2.0 g m^{-2}
21 yr^{-1} . In their study, both DIN and DON concentrations, measured in water samples
22 collected from plots characterised by similar moss composition, increased with
23 background N deposition but the latter increased more sharply. Baxter et al. (1992)
24 reported an accumulation of amino acids in mosses receiving NH_4NO_3 , which could be a
25 potential source of DON released into the water surrounding mosses.

26 The tracer application as $\text{NH}_4^{15}\text{NO}_3$ indicates that all ^{15}N -ammonium found in pore water
27 (Figs. 8 and 9) had to result from transformation of nitrate to ammonium. Such a process
28 can either take place in the soil by microbial reduction (Hemond 1983) or in the plant
29 tissue by an enzymatic reduction of assimilated nitrate (Guerrero et al. 1981). At Degerö
30 Stormyr, where only very little nitrate was present (Fig. 6), there was no potential for

1 nitrate reduction. In Cors Fochno and Whixall Moss cores the ^{15}N application increased
2 the amount of nitrate over the duration of the dry period (Fig. 6) and more $^{15}\text{N-NH}_4^+$ was
3 found at these sites as well (Fig. 8). The highest $^{15}\text{N-NH}_4^+$ excess was recorded at Lille
4 Vildmose, where also nitrate concentrations were high. In Frölichshaier Sattelmoor cores,
5 where nitrate concentrations reached 38 mg L^{-1} , most probably nitrate ammonification
6 took place since ammonium accumulated towards the end of the dry period (Fig. 6). The
7 lower isotopic signal recorded at the Frölichshaier Sattelmoor mesocosms could imply a
8 dilution effect caused by lower concentration of N in the applied solution in comparison
9 to the background N values.

10 The mechanisms of ammonium release may have changed with long-term N deposition,
11 as suggested by the distribution of the ^{15}N tracer (Fig. 9) in the mesocosms: At the sites
12 with low N inputs (DS), nitrate was apparently first assimilated by the plant layer and
13 later partly released as ammonium from the vascular plant roots. In these cores a direct
14 microbial transformation from nitrate to ammonium seems implausible, because nitrate
15 concentration levels were very low (Fig. 6). At the sites with elevated N inputs (FS) some
16 nitrate apparently bypassed the living moss layer (Fig. 8c) and was converted to
17 ammonium in the pore water. However, this pattern does not necessarily indicate a loss of
18 the *Sphagnum* filter. Even though the N content of *Sphagnum* tissue at Frölichshaier
19 Sattelmoor was at $10 \pm 3.8 \text{ mg g}^{-1}$ (Fig. 2) – the range encompassing the threshold N
20 concentration suggested by Lamers et al. (2000) to inhibit further N content increase –
21 the mosses did not suffer a substantial decline in biomass in the field and grew most of all
22 in the experiment (Supplementary Material, Fig. S3, Table S1).

24 **5 Conclusions**

25 Our results confirm that a legacy of elevated N deposition in ombrotrophic peatlands,
26 especially above levels of about $1.0 - 1.5 \text{ g N m}^{-2} \text{ yr}^{-1}$, leads to lower retention of N,
27 increased inorganic N concentrations in pore waters, and enhanced transfer of the
28 deposited N to shrubs. Higher inorganic and organic dissolved N concentrations and
29 more N in near-surface peat were found, and more of the ^{15}N label was recovered from
30 the aboveground shrub biomass at the polluted sites compared to the nearly pristine
31 Degerö Stormyr mire in Northern Sweden. As a consequence, a higher mobility of

1 dissolved N in peat is probable under more realistic hydrologic conditions than present in
2 our experiment. The shift to higher N mobility in the plant-peat soil system was,
3 however, more gradual and modest than we expected. It was not visibly linked to the
4 disappearance of *Sphagnum* mosses at the high long-term N deposition or to shifts in
5 species composition, if the somewhat limited information from our most polluted site at
6 the beginning of the 20th century is believed. Raised uptake of N by vascular plants at
7 sites exposed to higher long-term N deposition was accompanied by a *Sphagnum* N
8 absorption that remained substantial. Some former studies (Bubier et al. 2007;
9 Gunnarsson and Rydin 2000; Juutinen et al. 2010) alluded that the vitality of *Sphagnum*
10 suffers with increasing N saturation, which was undoubtedly the case in the mentioned
11 fertilization studies. Under conditions of our short-term experiments, however, the filter
12 function of mosses appeared to remain mostly intact, even when sites had been exposed
13 to long-term deposition levels considerably higher than 1.0 - 1.5 g N m⁻² yr⁻¹.

14

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3

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1 Tables

2 Table 1. Characteristics of the sampled peatlands.

Sites	Degerö Stormyr (DS)	Cors Fochno (CF)	Whixall Moss (WM)	Lille Vildmose (LV)	Frölichshaier Sattelmoor (FS)
Country	Sweden	Wales	England/Wales	Denmark	Germany
Lat (N)	64.181	52.502	52.915	56.846	51.791
Long (E)	19.559	-4.011	-2.771	10.183	10.531
MAP (mm)	602	1035	747	633	1222
MAT (°C)	1	9.7	8.7	7.6	5.2
MASL	277	4	91	8	765
Area (km ²)	6.5	10	9.48 ^y	20	0.18
Type	bor. mire	raised bog	raised bog	raised bog	raised bog
pH ^x	3.73±0.11	4.11±0.19	3.70±0.25	3.95±0.12	3.89±0.20
N deposition					
national models (g N m ⁻² yr ⁻¹)	<0.2	0.8	1.2	1.6	5.5-6.0
N deposition measured (g N m ⁻² yr ⁻¹)	0.2	n.a.	n.a.	1.7	4.7
N deposition IDEM (g N m ⁻² yr ⁻¹)	0.21	0.36	1.05	1.37	1.43
S deposition					
national models (g S m ⁻² yr ⁻¹)	0.3	0.21	0.23	0.65	5.5-6.0
Mean peat depth (m)	5	7	8	4-5	2.5-3.5
Water table level low/high (cm bsl)	17/7.5	15/0.5	23/2	31/1	30/2
Vegetation					
<i>Sph. capillifolium</i>		x	x		
<i>Sph. fallax</i>			x		

<i>Sph. magellanicum</i>					x	
<i>Sph. papillosum</i>						x
<i>Sph. pulchrum</i>		x				
<i>Sph. rubellum</i>	x					
<i>Andromeda polifolia</i>	x	x	x	x	x	x
<i>Calluna vulgaris</i>		x	x	x	x	X
<i>Erica tetralix</i>		x	x	x	x	
<i>Rubus chamaemorus</i>	x					
<i>Vaccinium oxycoccos</i>	x		x	x	x	X
<i>Eriophorum vaginatum</i>	x	x	x	x	x	X
<i>Eriophorum angustifolium</i>		x	x	x	x	

1 Site specific data – DS (Granberg et al., 2001; Sagerfors, 2007), CF, WM
2 <http://www.metoffice.gov.uk/climate/uk/averages/19712000/>, LV (Charman and Blundell, 2007), FS
3 (Baumann, 2009; Jensen, 1990). N and S deposition data (wet and dry deposition) – national models: DS
4 (Karlsson et al., 2012), CF and WM for year 2009 – Department for Environment Food and Rural Affairs
5 (DEFRA) accessed August 2012 at <http://pollutantdeposition.defra.gov.uk/data>, LV for year 2004
6 (Ellermann, et al. 2004), FS for 1993 (Gauger, 2009), – measured values: DS for 1990-1992 (Granberg, et
7 al. 2001), CF and WM data not available, LV (Ellermann et al., 2004), FS (Böhlmann, et al. 2005), – IDEM
8 model (Pieterse et al. 2007). Abbreviations: Lat – latitude, Long – longitude, MAP – mean annual
9 precipitation, MAT – mean annual temperature, MASL – meters above sea level, bsl – below sea level,
10 IDEM – Integrated Deposition Model (Bleeker et al., 2004). ^y The area of Fenn's, Whixall and Bettisfield
11 Mosses.

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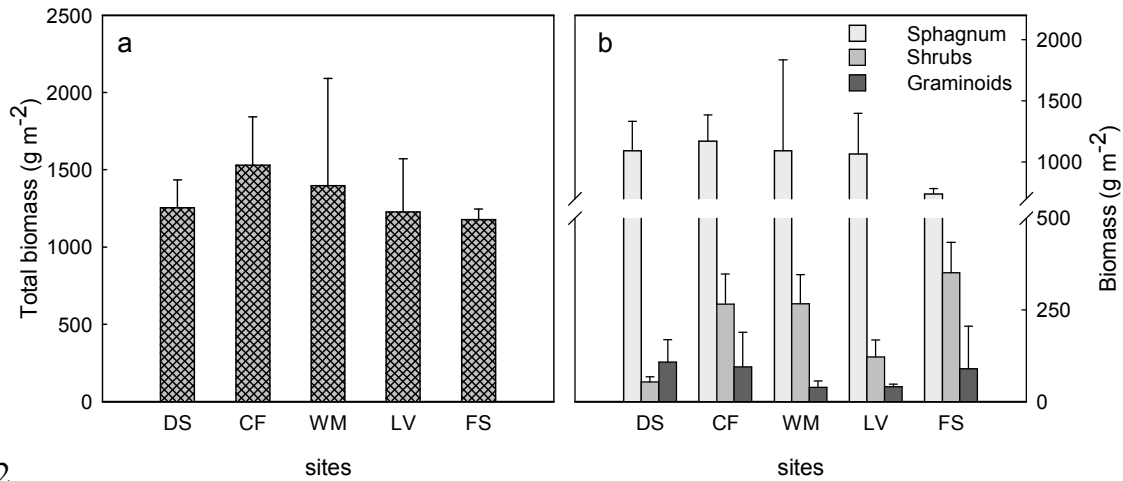
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1 Table 2. Recovery of ^{15}N (%) for all sites and percent contribution of N ecosystem
 2 pools to the total recovery at each site after 24 weeks of tracer application. NO_3^-
 3 and NH_4^+ were analyzed in peat pore water. Standard deviations are given in
 4 parenthesis.

Site	NO_3^-	NH_4^+	<i>Sphagnum</i> (0 - 5 cm)	Shrubs	Graminoids	Peat	Total recovery
Degerö	0.011	0.003	65.6	3.8	1.8	59.8	131.0
Stormyr	(0.002)	(0.003)	(22.6)	(1.0)	(0.5)	(45.8)	(25.3)
Cors	0.043	0.163	40.4	6.2	0.8	57.0	104.7
Fochno	(0.045)	(0.030)	(15.0)	(3.4)	(0.3)	(18.7)	(12.0)
Whixall	0.014	0.284	23.6	5.4	0.4	68.9	98.7
Moss	(0.002)	(0.134)	(9.5)	(3.3)	(0.4)	(7.2)	(6.3)
Lille	0.031	0.661	57.8	5.7	1.4	21.9	87.5
Vildmose	(0.025)	(0.282)	(23.7)	(5.1)	(1.2)	(20.9)	(16.2)
Frölichshaier	0.051	0.374	45.5	12.4	1.2	36.2	95.7
Sattelmoor	(0.037)	(0.065)	(6.3)	(4.3)	(1.6)	(12.8)	(6.5)

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1 **FIGURES**

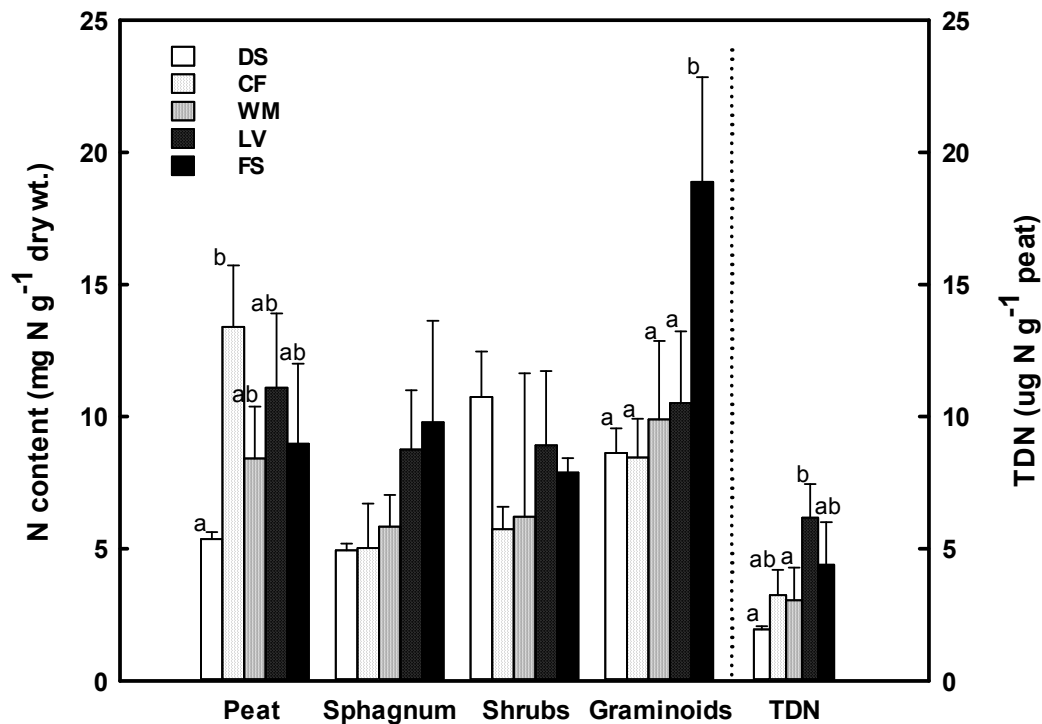


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4 Figure 1. Total biomass (A) and the biomass of the plant functional types (B) (means ± SD). Site
5 abbreviations: DS – Degerö Stormyr, CF – Cors Fochno, WM – Whixall Moss, LV – Lille
6 Vildmose, FS – Frölichshaier Sattelmoor.

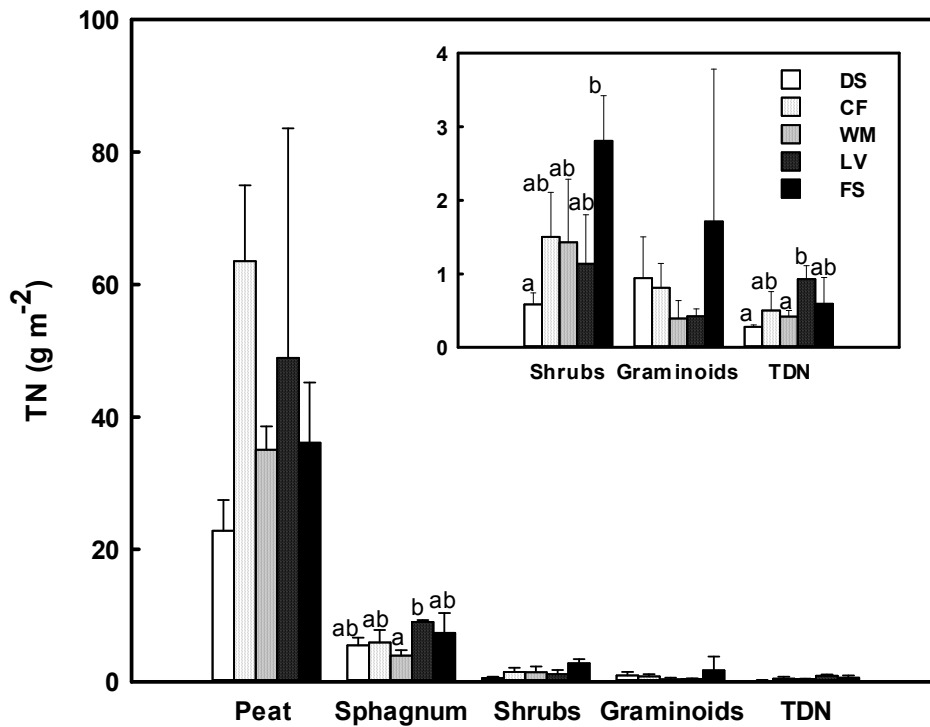
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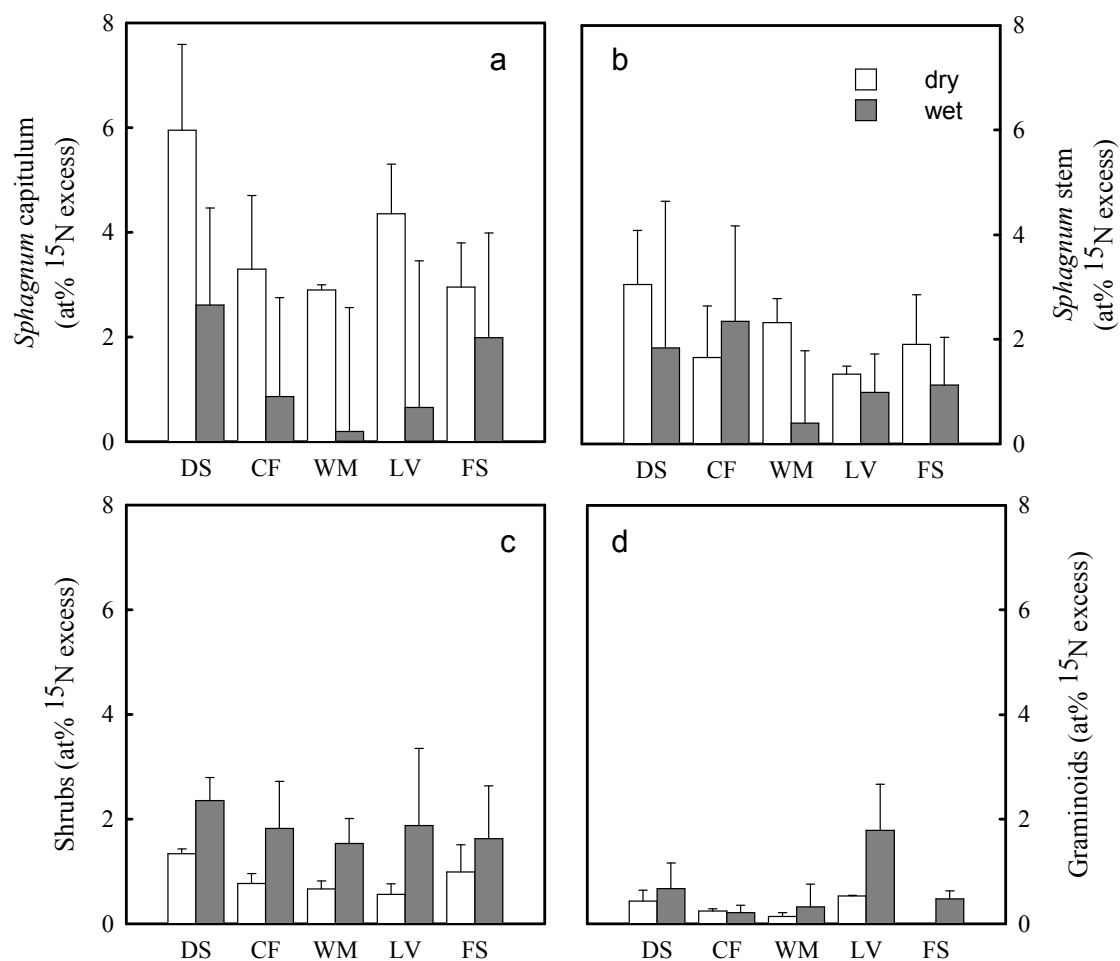
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Figure 2. Nitrogen content (mean \pm SD, $n = 3$) of peat (5-15 cm depth), *Sphagnum* (0 - 5 cm below surface), shrub, graminoid and total dissolved nitrogen (TDN) (0-15 cm depth) pools at the end of the experiment. Letters *a* and *b* - significant differences in the mean values of the sites at each pool (one-way ANOVA, Tukey Test). A bar without a letter - no significant difference. Site abbreviations: DS – Degerö Stormyr, CF - Cors Fochno, WM - Whixall Moss, LV – Lille Vildmose, FS – Frölichshaier Sattelmoor.



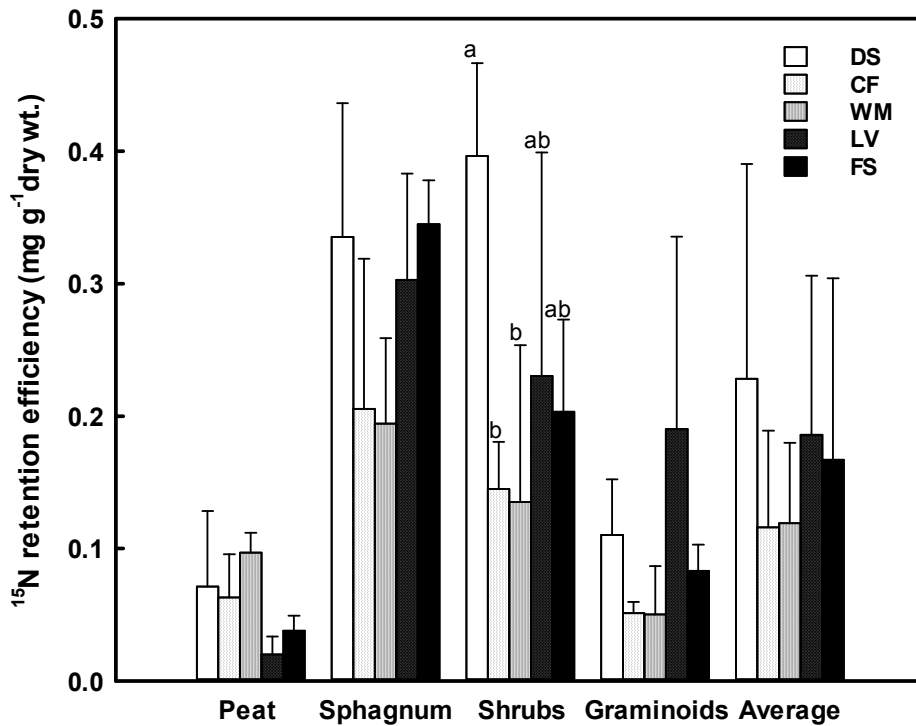
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Figure 3. Total nitrogen content (mean \pm SD, n = 3) in the ecosystem pools investigated standardized to the peatland surface area. The *Sphagnum* pool contained mass from 0 to 5 cm below surface. Letters *a* and *b* - significant differences in the mean values of the sites at each pool (one-way ANOVA, Tukey Test). A bar without a letter - no significant difference. Site abbreviations as in Figure 1.



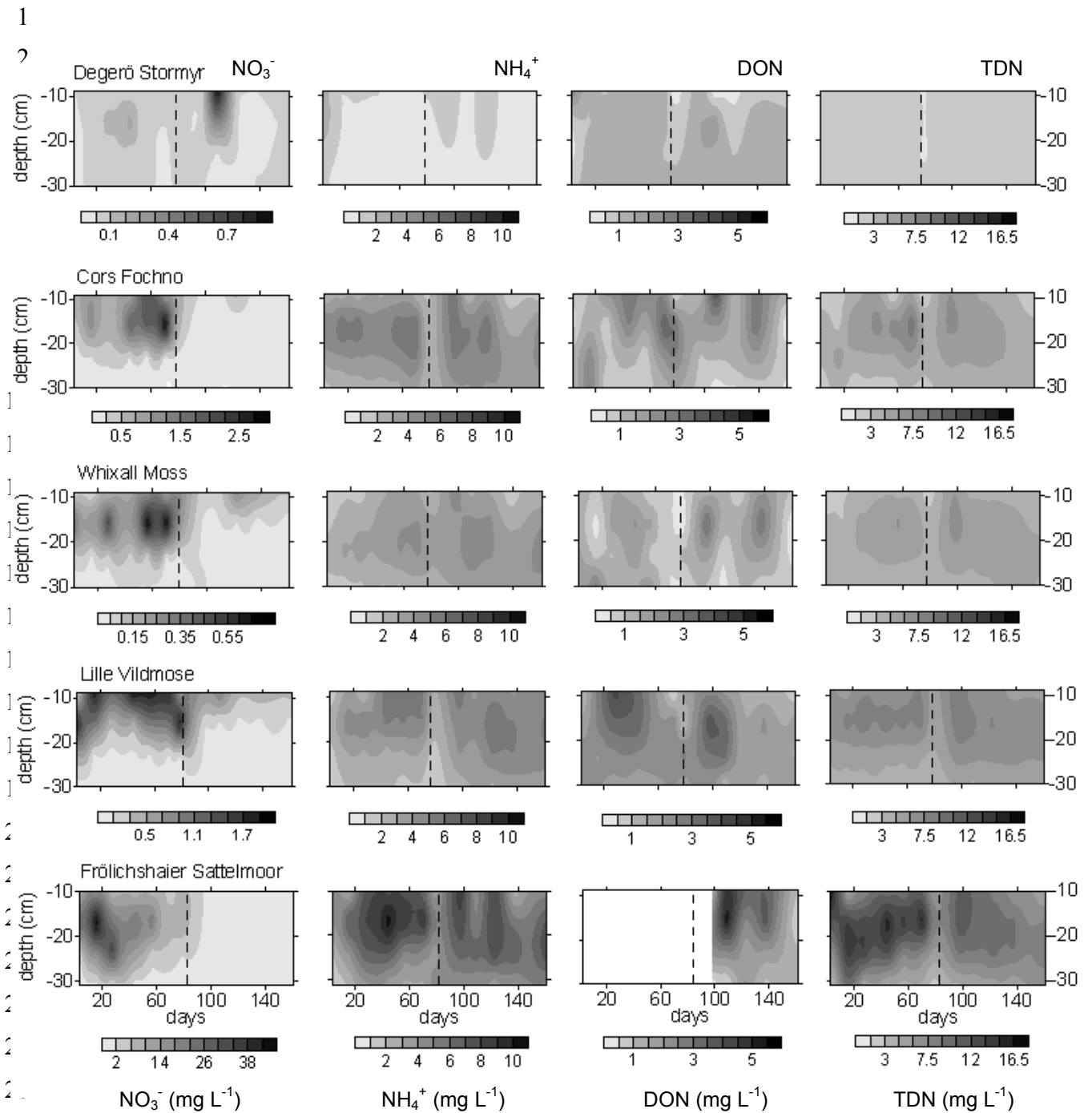
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 2 Figure 4. ^{15}N abundance (mean \pm SD, n = 3) expressed as at% ^{15}N excess in vegetation
 3 during dry (dry) and during wet (wet) period. A – *Sphagnum capitulum*, B – *Sphagnum*
 4 stem, C – shrubs, D – graminoids. One-way ANCOVA indicated a significant effect of N
 5 deposition for *Sphagnum capitulum*, graminoids, and of the dry and wet period for
 6 *Sphagnum capitulum* and shrubs; a Tukey post-hoc test failed to identify significant
 7 differences for individual treatments, however. Site abbreviations as in Figure 2.

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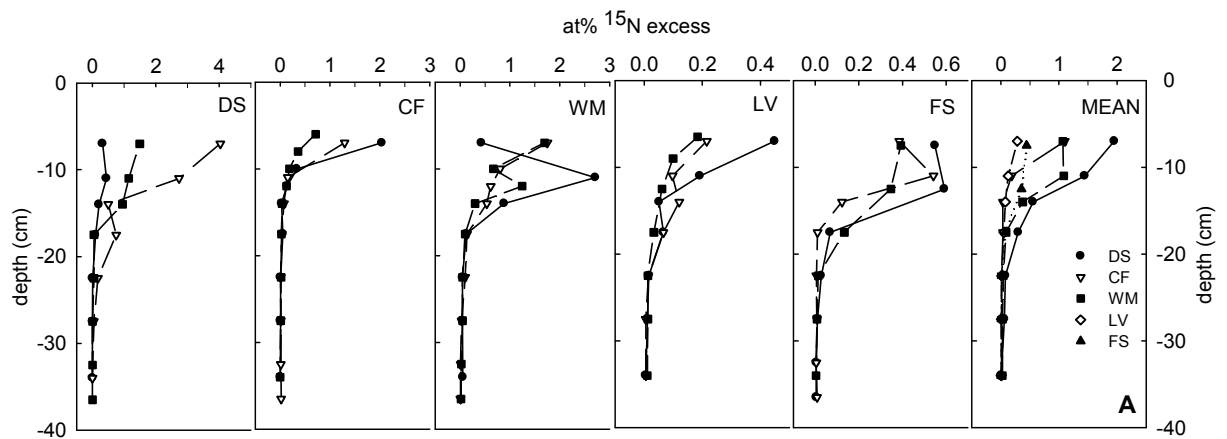
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Figure 5. ¹⁵N retention efficiency (mean ± SD, n = 3) of ecosystem pools, calculated by dividing retained ¹⁵N in each pool at the end of the experiment by dry mass of the corresponding pool. 'Average' represents the mean ¹⁵N retention efficiency of all pools (mean ± SD, n = 4). Letters *a* and *b* - significant differences in the mean values of the sites at each pool (one-way ANOVA, Tukey Test). A bar without a letter - no significant difference. Site abbreviations as in Figure 1.

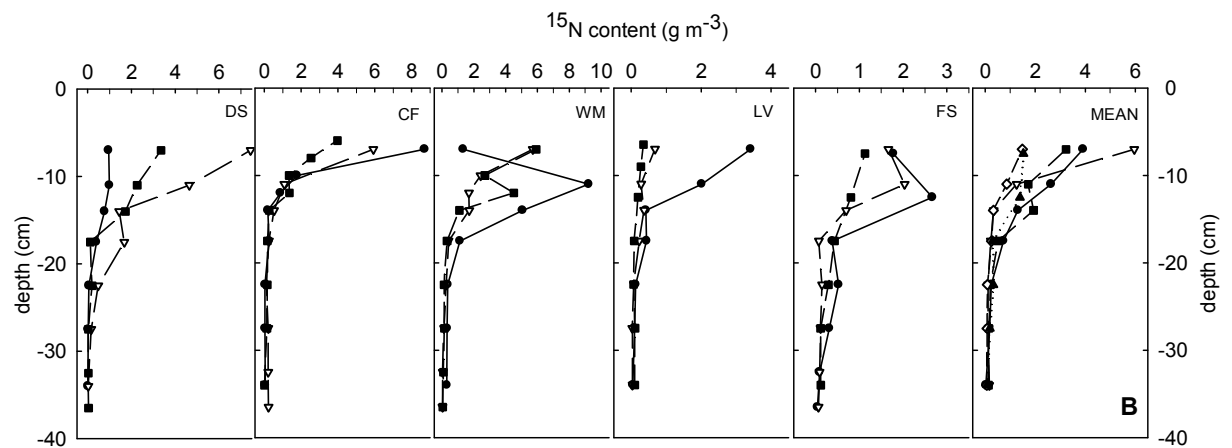


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 28 Figure 6. Dissolved nitrogen concentrations in the pore water: nitrate (NO_3^-), ammonium
 29 (NH_4^+), dissolved organic nitrogen (DON) and total dissolved nitrogen (TDN) Please
 30 note the varying scale bars for NO_3^- . Vertical dashed lines mark the transition between
 31 dry (left) and wet (right) period.

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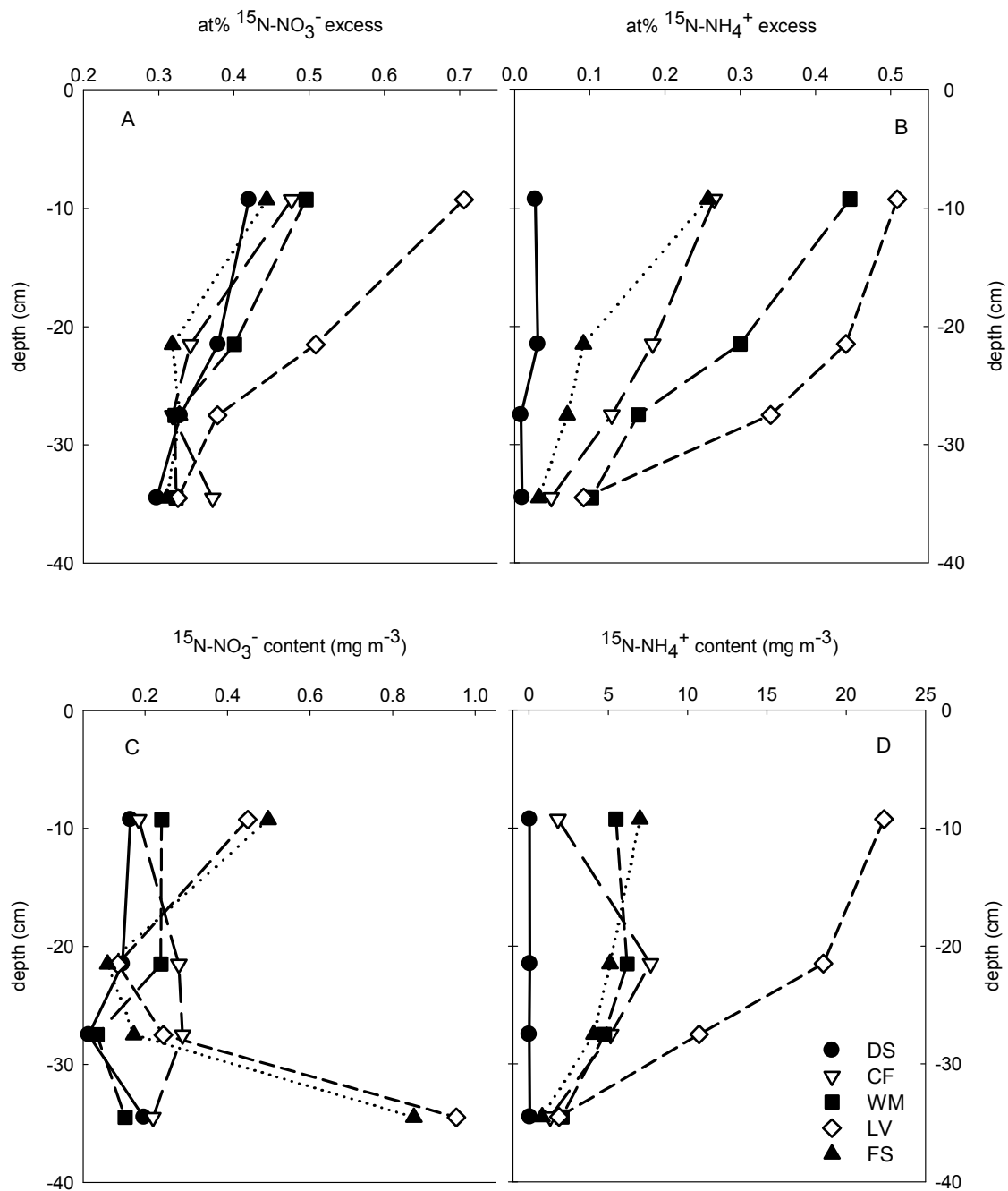


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5 Figure 7. Depth profiles of peat at% ^{15}N excess (A) and peat ^{15}N content (B) for three
6 replicate cores from each site (6-9 samples/core) and the mean at% ^{15}N excess and peat
7 ^{15}N content for each site. The scale of the X-axes varies across panels. Site abbreviations
8 as in Figure 1.

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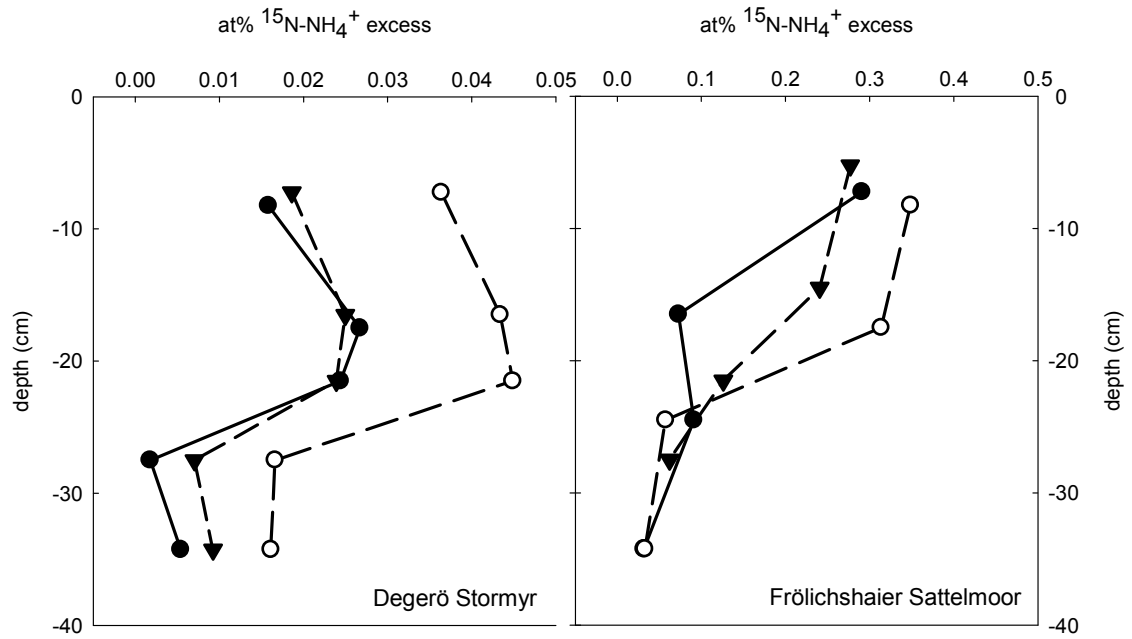
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4 Figure 8. Pore water ^{15}N excess and ^{15}N content: (A) ^{15}N -nitrate (at% excess), (B) ^{15}N -
 5 ammonium (at% excess), (C) ^{15}N -nitrate (mg m^{-3}), (D) ^{15}N -ammonium (mg m^{-3}). The
 6 scale of the X-axes varies across panels.

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4 Figure 9. Atom % ¹⁵N-NH₄⁺ excess in pore water of three replicates from Degerö
5 Stormyr (left) and Frölichshaier Sattelmoor (right). Each symbol in the profile represents
6 the middle depth of the section from which the water was extracted. The scale of the X-
7 axes varies across panels.

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