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Spatio-temporal trends of nitrogen deposition and climate effects on *Sphagnum* productivity in European peatlands

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Abstract

To quantify potential nitrogen (N) deposition impacts on peatland carbon (C) uptake, we explored temporal and spatial trends in N deposition and climate impacts on the production of the key peat forming functional group (*Sphagnum* mosses) across European peatlands for the period1900 - 2050. Using a modelling approach we estimated that between 1900-1950 N deposition impacts remained limited irrespective of geographical position. Between 1950-2000 N deposition depressed production between 0-25% relative to 1900, particularly in temperate regions. Future scenarios indicate this trend will continue and become more pronounced with climate warming. At the European scale, the consequences for *Sphagnum* net C-uptake remained small relative to 1900 due to the low peatland cover in high-N areas. The predicted impacts of likely changes in N deposition on *Sphagnum* productivity appeared to be less than those of climate. Nevertheless, current critical loads for peatlands are likely to hold under a future climate.

Keywords: air pollution, carbon balance, critical load, ecosystem change, peat mosses

Capsule: Temperate *Sphagnum* peatlands are vulnerable to current and future N deposition and current critical loads for peatlands are likely to hold under a future climate.

Introduction

Deposition of reactive nitrogen (N) has increased steadily in many parts of the world since the industrial revolution (Galloway *et al.* 2003). At present N deposition levels are either stabilizing at a high level (Europe) or continue to increase in other parts of the world (de Vries & Posch 2011). N effects are numerous, ranging from biodiversity loss to increased leaching of nitrate to aquifers and lakes (e.g. Bergström & Jansson 2006). To improve risk assessments and support policy decisions, critical N deposition loads have been defined for many ecosystems (Bobbink & Hettelingh 2011), including *Sphagnum*-dominated peatlands. These extremely nutrient-poor ecosystems are among those most sensitive to N enrichment, and show changes in species composition above N deposition levels of 5-10 kg ha^{-1} yr^{-1} (Bobbink & Hettelingh 2010). Recent work suggests that N-impacts on *Sphagnum* depends on climatic factors (Heijmans *et al.* 2008; Limpens *et al.* 2011), implying critical loads may vary across geographical gradients and may change with climate warming. Our study is the first attempt to explore the spatial distribution of N effects on *Sphagnum* production in peatlands for past, present and future.

Sphagnum-dominated peatlands cover large areas of the boreal-temperate zone and store a substantial amount of the global soil carbon (C) pool (Rydin & Jeglum 2013). Peat mosses (genus *Sphagnum*) play a key role in these ecosystems as they efficiently engineer an environment that facilitates their own growth while being hostile to vascular plants (van Breemen 1995). Their unique properties are both directly and indirectly responsible for the long-term C sequestration of northern peatlands. *Sphagnum* directly affects C-sequestration by producing litter that decays at a slower rate than vascular plants (Freeman *et al.* 2001), thus forming a much greater proportion of the peat than expected from their primary production (Wallén, 1992),. *Sphagnum* litter/peat is also responsible for the wet and acidic environment that suppresses decay, thus indirectly facilitating long-term C-sequestration. Thus, *Sphagnum* production and abundance are of vital importance for the functioning and C accumulation of *Sphagnum*-peatlands (Johnson & Damman 1993). The proportion of annual *Sphagnum* production that is eventually integrated into long-term C storage (peat accumulation) varies between 0-10%, depending on environmental conditions (Rydin & Jeglum 2013). This can translate into an average carbon accumulation of circa 20 g C m^{-2} yr^{-1} (Roulet *et al.* 2007). Although peatland habitats are usually resilient to environmental change (Belyea & Baird 2006), nutrient alterations can lead to drastic changes in the composition of the vegetation. In general vascular plants benefit from the surplus N and become more dominant while the growth ofpeat mosses decreases due to litter burial and shading (Berendse *et al.* 2001). There is also experimental evidence of more direct effects of increased N availability on *Sphagnum* performance, such as increased photosynthesis, P limitation and even toxicity at very high N levels (Granath *et al.* 2009, Granath *et al.* 2012, Limpens and Berendse 2003). Consequently, N deposition has been suggested as a potential threat to C sequestration of boreal and temperate peatlands (Gunnarsson *et al.* 2008; Dise 2009). Yet, attempts to quantify this threat are lacking. This is in sharp contrast to forest ecosystems where N deposition effects on C sequestration have been quantified at a European scale (de Vries *et al.* 2006; de Vries & Posch 2011).

A modelling approach is necessary to quantify the effects of N deposition at a larger scale, and until recently, no model was available for *Sphagnum*. A recent meta-regression analysis (Limpens *et al.* 2011), was an important step forward in the process of understanding and quantifying the effect of N on *Sphagnum* growth in peatlands. Using results of N application experiments across the northern hemisphere, the authors identified some of the variables that may affect *Sphagnum* growth response to N deposition. This analysis resulted in a statistical model that describes the impact of N deposition on *Sphagnum* production in interaction with climatic factors. By combining this statistical model with N deposition and climate data for European peatlands for the past, present and future (1900-2050) we investigated, i) how N deposition has affected *Sphagnum* production, temporally and spatially across Europe, over the 20th century, ii) the effect of future N deposition on *Sphagnum* production, given future climatic and N deposition projections, and iii) the overall consequences for *Sphagnum* production and potential C-uptake at European scale.

Methods and Materials

Modelling the effect of nitrogen deposition on Sphagnum

We adapted the meta-regression model of Limpens et al (2011), further referred to as Limpens-Granath model, to model the effect of N deposition on *Sphagnum* productivity under different climatic conditions. Our adapted model is expressed as:

$$\log_e (P_{treat} / P_{control}) = \mu + a \times N_c + b \times N_{add} + c \times T_j + d \times P_a + C + Error \qquad (1)$$

where μ is the intercept, N_c is the current mean annual N deposition at the site at the time of treatment, N_{add} is the annual added N at treatment plots, T_j is the mean July temperature (which is strongly correlated with mean summer temperature, June-August), P_a is mean annual precipitation, $P_{control}$ is the *Sphagnum* productivity at the control plots receiving only N_c, and P_{treat} is the *Sphagnum* productivity at the treatment plots, receiving $N_c + N_{add}$. C and a, b, c, d are regression coefficients, where C also includes the effects of phosphorus application and presence of vascular plants. The *Error* term captures the unexplained between-experiment variation.

Adaptations to the Limpens-Granath model included the use of more recent climate and background N deposition data, and a further simplification of parameters and adaptation of N_c

and N_{add} to make the model suitable for comparing N effects over different time periods instead of between treatments. These adaptations are discussed below.

Parameterization

The model was parameterized using the same experimental data as in Limpens et al. (2011) The most important details are given in Table S1 (sources, location, species, N application rate). In contrast to Limpens et al. (2011), we used more recent climate and background N deposition data. For the adapted model (1) we used gridded average climate data for a 10-year period encompassing the timing of the experiments based on the Climate Research Unit (CRU) data base (Mitchell *et al.* 2004). Furthermore, annual total background N deposition (i.e. wet plus dry) was used instead of only wet deposition as in Limpens *et al.* (2011). Most N application experiments were conducted in Europe and wet and dry N depositions for the years of the experimental duration were extracted from the European Monitoring and Evaluation Programme (EMEP MSC-W model results, http://www.emep.int/mscw/index_mscw.html). For non-European sites, N deposition was retrieved from original publications. More details on N deposition and climate data can be found in the methods section below. The model was fitted using a Bayesian approach in the R package MCMCglmm (Hadfield 2010) to more easily include parameter uncertainties in the spatial models. For parameterization, we ran the model for 140 000 iterations after a burn-in of 30 000 iterations, using non-informative priors. Changing the priors did not alter the results and ensures little influence of the priors on the posterior (see Limpens *et al.* (2011) for details on fitting hierarchical meta-regression models accounting for within-study dependence).

The Limpens-Granath model included the effect of microhabitat (moist lawn vs. dry hummocks) and an interaction between microhabitat and temperature. As the proportions of microhabitats in peatlands are not well-documented, we removed the terms involving microhabitat in the model, generalizing our predictions. In addition, the Limpens-Granath model included variables that were manipulated in the experiments: phosphorus application and presence of vascular plants (some experiments removed vascular plants). In our predictions we assumed that phosphorus was not added and vascular plants were not removed.

To test if the adapted Limpens-Granath model would lead to a loss of predictive value, we compared both the original and adapted models using the experiment data set of Limpens *et al*. (2011). Our simpler model performed as well as the original model, with predictors explaining 51% (present model) and 53% (Limpens-Granath model) of the variation among experiment outcomes (log response ratio), respectively. Model parameters estimated for our revised model are given in Table 1.

Our model was applied on a 0.5×0.5 degree longitude-latitude grid (8539 grid cells with peatlands) covering Europe except for the most eastern parts where the peatland distribution is not available (Ukraine, Belarus, Moldova and Greece, see Figure 2).To make spatial predictions, we used July temperature, annual precipitation and annual N deposition data for each grid cell. Model uncertainty (parameter uncertainty and random variation) was incorporated by performing random draws of parameter estimates from the joint distribution of the meta-regression model. We made 5000 random draws for predicting the outcome at each grid cell and time period. From these 5000 predictions we calculated the mean and the 90% and 50% uncertainty intervals.

Including all model uncertainties gave wide uncertainty intervals. A large part of that uncertainty comes from the unexplained between-study variation (Table 1), which is mainly a result of large within-peatland variation in *Sphagnum* species and vascular plant cover. Since we were not interested in the within-peatland variation but rather in the aggregated response of the whole peatland to N deposition, we removed the uncertainty related to *Sphagnum* species for the uncertainty graphs. To this end we first analyzed a subset of the data used in Limpens *et al.* (2011) that only contains studies with more than one species (see Table S1). From this data we estimated that the species component explained about 25% of between-study variation. This variation was removed when prediction intervals were created for the graphs presented in the supporting information (see figures in Appendix S1 in Supporting Information).

To summarize the effect over Europe we calculated weighted means of predictions using proportions of peatlands in each grid cell as weights. The estimation of the peatland cover is described later in the methods section. Effects on carbon uptake were calculated by assuming a mean annual *Sphagnum* production of 200 g dry matter m^{-2} yr^{-1}. This value is derived from a meta-analysis of *Sphagnum* production (Gunnarsson 2005) enhanced by the production rates observed in the control plots of the N addition experiments that formed the basis of our model. The 200 g m^{-2} yr^{-1} is a representative, albeit conservative (e.g. Wieder *et al.* 2010), estimate of long-term production values and thus highly suitable for the 50-year time step adopted in our study. To convert the dry matter production to C-uptake we assumed a dry tissue carbon content of 50% (Rydin & Jeglum 2013).

Modelled time periods and scenarios

We analysed N deposition effects between 1900-1950, 1950-2000 and four different scenarios for the period 2000-2050; combining (1) two N deposition scenarios (current N deposition, using means of 2000-2010, and future N deposition, according to the Global Energy Assessment current legislation, GEA-CLE), with (2) two climate scenarios (current climate, using means of 1950-2000, and future climate, according to the A1 climate scenario). For more information on these scenarios see sub-section *Nitrogen deposition 1900-2050* and *Climate data 1900-2050.*

Modelling N effects over time

The response variable in the Limpens-Granath model was the ratio of *Sphagnum* productivity in treatment over control, i.e. the model predicts the *change* in *Sphagnum* productivity compared to a reference point. Consequently, to apply the Limpens-Granath model based on N addition experiments to field conditions, we needed to set a reference point with which to compare the effects of increasing N deposition. We used the year 1900 as reference point, assuming that N deposition at that time reflects pre-industrial times (de Vries & Posch 2011).

N load in the Limpens-Granath model was divided over N deposition at the experimental site (N current = N_c) and experimentally applied N (N_{add}). For our adapted model we also divided N deposition over the terms N_c and N_{add}, where N_c presented the impact of long-term historic N deposition, and N_{add} the impact of additional N deposition. We used one or two allocation steps, depending on the change in N deposition between 1900 and 2050. If N deposition changed less than 0.4 g m^{-2} between 1900 and 2050, no reliable predictions could be made between the periods as the Limpens-Granath model assumes a minimum N_{add} of 0.4 g m^{-2}. One allocation step was used when the change in mean N deposition between 1900-2050 exceeded 0.4 g m^{-2} yr^{-1}. Then the N deposition in 1900 was allocated to N_c, and all other N to N_{add}. If the change in N deposition between 1950-2000 to 2000-2050 was higher than 0.4 g m^{-}

^{2} yr^{-1} we used two allocation steps: N deposition in 1900 was allocated to N_c for the period 1950 to 2000, and the average N deposition between 1950 and 2000 was allocated to N_c for the period 2000-2050. In the latter case, the outcome was multiplied with the first outcome (effect 1900 to 1950-2000) to keep comparisons to 1900.

Model evaluation

The sensitivity of our model was evaluated in two ways; 1) to investigate the effect of potential uncertainty of our modelled predictors we ran the model with an uncertainty (standard deviation set to 5% of the value) around modelled predictor values (N deposition, temperature and precipitation), and, 2) to test the effect of different ways of allocating N deposition over N_c and N_{add}. In addition, we explored an alternative model where the parameters N_c and N_{add} were merged into a single parameter (total N addition, N_{c+add}). The test of this alternative model was also restricted to the 0.4 g m^{-2} yr^{-1} minimum increase since 1900.

Nitrogen deposition 1900-2050

Oxidised and reduced N deposition were calculated with the atmospheric transport model of EMEP/MSC-W (Simpson et al. 2012). Historic NO_x and NH_3 emissions were taken from Lamarque *et al.* (2010). Predictions for the period 2000-2050 were based on a (1) constant N deposition scenario, i.e. fixing N deposition to the current levels, here defined as the mean for the period 2000-2010, and, (2) Global Energy Assessment (GEA) current legislation scenario (GEA-CLE). The GEA-CLE deposition scenario assumes full implementation of all current and planned air pollution legislation world-wide until 2030.

Climate data 1900-2050

Historical meteorological data were taken from a high resolution European data base (Mitchell *et al.* 2004) that contains monthly values of temperature, precipitation and cloudiness for the years 1901–2000 for land-based grid-cells west of 32ºE of size 10'×10' (approx. 15 km×18 km in central Europe). In the simulations, 10-year averages were taken centred around 1900, 1910, …, 1990 (1900 is the average of 1901–1905; 1910 of 1906–1915 etc.) to smooth the climate pattern. For future values we used two climate scenarios: (1) constant climate after 1990 (using the average of the 1961-1990 period); (2) from 2010 onwards the A1 storyline from the Special Report on Emission Scenarios (SRES) with its related climate scenario (Nakićenović & Swart 2000). This scenario, which is from the IPCC scenario family with the highest emissions, reflects a future world with globalization and rapid economic growth, low population growth, and the rapid introduction of new and more efficient technologies everywhere. The A1 scenario was chosen in this study as the most likely future scenario, considering that current CO_2 emission trends exceed this most pessimistic IPCC scenario (Raupach *et al*. 2007). Mitchell *et al.* (2004) used results from the HADCM3 General Circulation Model to derive monthly meteorological data for the years 2001–2100 from this (and several other) scenario, which we again averaged over 10-year periods for use in the simulations. For the transition period, 1990–2010, data were interpolated linearly. Under the A1 climate scenario, temperatures over Europe increase by about 3±1ºC between 1990 and 2050, whereas median precipitation hardly changes, with precipitation at southern sites decreasing and at northern sites increasing compared to 1990 (see also de Vries & Posch 2011).

Spatial distribution of peatlands in Europe

We improved the estimation of the coverage of open *Sphagnum* dominated peatlands by integrating the CORINE (CO-oRdination of INformation on the Environment) land cover 2006 of the European Environmental Agency (EEA) with the Forest Map of the Joint Research Centre (JRC) of the European Commission. The CORINE land cover is derived from high resolution satellite data (e.g. Landsat-TM) by computer assisted visual interpretation in combination with ancillary data. The minimum mapping unit is 25 ha and for line elements the minimum width is 100 m. Büttner and Maucha (2006) validated the CLC2000 and found a total reliability of CLC2000 was 87.0 ± 0.8%. The JRC Forest Cover Map is a 25 m spatial resolution raster Pan-European Forest / Non Forest Map with target years 2000 and 2006 derived from high resolution satellite data such as LISS III and SPOT 4/5 imagery. In the JRC forest map, forests are defined as vegetation dominated by trees, with canopy closure more than 30% and trees higher than 5 m. For its integration with CORINE, the JRC Forest Map was resampled to a 100 m grid (same resolution as CORINE) using a majority filter.

From CORINE land cover we selected the land cover class 4.1.2 Peat bogs, which has been defined as: peatland consisting mainly of decomposed moss and vegetable matter which might be exploited (Bossard et al. 2000). It includes minerogenic peatlands (fed by ground water) with mosses (mostly *Sphagnum*), ombrogenic peatlands (fed only by precipitation and dominated by *Sphagnum*) such as boreal bogs, boreal bogs with reticulated structure (aapa), blanket bogs, and fossil arctic peat bogs (palsa). The integrated map of JRC forest and CORINE land cover (2006), focusing on non-forested peatlands, was analysed over European sites where N application experiments used in the meta-analysis were performed, and over other sites where *Sphagnum* production had been recorded in the literature. Our combined map showed an improvement over CORINE 2006 concerning open, *Sphagnum* dominated peatlands.

The gridded peatland distribution map was first re-projected and then overlaid with the gridded climate/N deposition map (0.5×0.5 longitude-latitude grid) to calculate the proportion of peatland per cell. This was done using the R packages *raster* (Hijmans & van Etten 2012) and *rgdal* (Keitt *et al.* 2012). All maps are shown with the EPSG:3035 projection.

Results

N deposition trend and peatland cover

Estimated N deposition varied over time and space. At the beginning of the 1900s, N deposition was generally low, except for a region in central Europe and the southern British Isles. Between 1900 and 1950 N deposition increased slightly, but not more than 0.4 g m^{-2} yr^{-1} on average over the whole period (Fig 1a). From 1950 onwards, N deposition rose quickly, reaching peak values around 1980-1990, after which it decreased slightly and levelled off to the present levels in many areas. The total change in N deposition exceeded 0.4 g m^{-2} yr^{-1} in most areas (Fig 1b). Predictions for 2000-2050 under the current legislation N emission scenario (GEA-CLE) vary per region, with N deposition increasing or decreasing, depending on location (Fig 1c). In the constant (mean 2000-2010) scenario, most areas showed a minor decrease compared to the 1950-2000 period (not shown).

Combining CORINE land cover data and the JRC forest map, we estimated open/semi-open *Sphagnum*-dominated peatland cover in Europe to be10.5 million ha. About 20% of these peatlands are defined as forested. The highest cover of peatlands is found in Fennoscandia and the British Isles (Fig 2).

Spatial predictions for Sphagnum productivity

For the period 1900-1950 we could not make reliable predictions for changes in *Sphagnum* productivity as the change in N deposition remained below 0.4 g m^{-2} (the minimum amount needed for modelling predictions, see Methods) for all regions. For 1950-2000, large areas of Europe did show a sufficient increase in N deposition for making predictions. This area includes approximately 1 million hectares of *Sphagnum* dominated peatlands. Over this period, our model estimated that N deposition depressed *Sphagnum* production between between 0 and 25% in western, central and eastern Europe (Fig 3a), but increased production in south-eastern Sweden. N deposition in parts of Ireland and northern Fennoscandia did not change enough to allow reliable predictions. When summarized over all predictions and taking peatland cover into account, the overall N effect on *Sphagnum* production, and consequently on the *Sphagnum* C balance of the living parts, was close to zero (Table 2).

Over 2000-2050 N deposition is predicted to depress *Sphagnum* production further under both the constant N emission scenario (using average N deposition of 2000-2010; Fig 3c) and the current legislation N emission scenario (GEA-CLE deposition scenario; Fig 3b). Without a change in climate however, predictions are similar to 1950-2000 (Fig. 3d). The strongest overall reduction of *Sphagnum* production occurs under the constant N deposition and A1 climate scenario (Table 2, Fig 3c) and the weakest is under the reduced N deposition and constant climate scenario (Table 2, Fig 3d). The predicted impacts of N deposition appeared to be less (on average 3 percentage points difference between constant and reduced N deposition) than those of climate change (on average 7-8 percentage points difference between constant climate and climate change according to A1; see table 2).The consequences of the different scenarios for C uptake show large spatial variation, but little-no difference when upscaled to Europe (Table 2).

Model uncertainty and sensitivity

Including all model uncertainties gave wide uncertainty intervals even when correcting for contribution of within peatland uncertainty. Hence parameter uncertainty is the largest contributor to the overall uncertainty (Supporting information, Fig. S1, S2).

When looking at the sensitivity of the model, a standard deviation of 5% for a predictor value (e.g. an N deposition value of 1.0 g m^{-2} yr^{-1} is assumed to be estimated with a standard deviation of 0.05 g m^{-2} yr^{-1}) did not change our predictions substantially. If we did not use allocation steps for N_c, but only one fixed N_c (N deposition in1900) for all time periods, our future predictions (2000-2050) suggested less negative effects of N deposition. Also, the model where N_c and N_{add} were merged into one parameter (N_{c+add}) did not diverge much from our original model. In this case, however, the model produced more extreme predictions near the boundaries of model space and stronger negative effects were predicted at lower N deposition levels because a larger proportion of N deposition was allocated to N_c. The instability of this model (N_{c+add}) makes it less useful despite its simplicity and thus the results from the two-step allocation model are the only ones presented here.

Discussion

Nitrogen deposition impacts

For the second half of the 20th century, our model suggests that N deposition will depress *Sphagnum* production in many areas by 0-25% relative to 1900. This reduction nearly equals the year-to-year variation due to weather conditions observed in contemporary *Sphagnum* production studies (e.g. Vitt 1990). These modelled trends correspond to independent field observations of peat accumulation (Gunnarsson *et al.* 2008), vegetation changes (Kapfer *et al.* 2011) and *Sphagnum* N concentration (Malmer & Wallén 2005).

The N deposition impact varied spatially, with N deposition depressing *Sphagnum* production mostly in peatlands approximately south of 60° latitude, with the exception of Eastern Sweden. North of 60° latitude, changes in *Sphagnum* production could generally not be modelled because N deposition changed less than 0.4 g m^{-2} yr^{-1} relative to 1900. However based on our model, it is likely that N deposition had little effect on *Sphagnum* production and probably stimulated growth. Unfortunately, there are no long-term data sets on *Sphagnum* production that we can use for direct validation of our model results, restricting validation to comparisons of patterns observed in vegetation composition and litter production which may be viewed as indirect proxies for *Sphagnum* production. Below we compare our results with patterns observed in vegetation composition and litter production in bogs. Our spatial distribution of N deposition impacts corresponds with that observed in studies focussing on peatland vegetation composition. For example, in Southern Sweden (60° latitude) where our model estimates a net positive effect over the 20th century, Gunnarsson et al. (2000) found no evidence of N-induced changes in vegetation composition, whereas in England where our model estimates negative effects, Chapman & Rose (1991) reported negative effects on vegetation, notably on *Sphagnum* cover. Another interesting example is the peatland Store mosse in SW Sweden. Malmer & Wallén (2004) estimated that about 50% less litter (*Sphagnum* and vascular plants) was produced in the 20th century compared to the 19th century. A direct comparison with our estimate (20-25% reduction) is not straight forward since we focus on *Sphagnum* and single out the effect of N deposition. In their study, Malmer & Wallén (2004) acknowledge that N deposition is a likely important factor causing the decrease of litter from *Sphagnum*, together with general drier conditions. Although being a single peatland, this gives some quantitative support for our predictions that needs to be scrutinized further. The scarcity of suitable validation data highlights the need of long-term monitoring of processes, such as productivity, instead of only state variables, such as vegetation composition.

Our future scenarios suggest that N deposition impacts predicted for the period between 1950-2000 will continue in the 21st century, becoming more pronounced with climate warming. At the European scale, however, the net N deposition impact on carbon uptake by *Sphagnum* remains small due to the low peatland cover in high N deposition regions: ranging between -6 to -15 g C m^{-2} yr^{-1} relative to 1900 for the period 1950-2050, depending on N deposition scenario and climate warming. However, at the peatland scale N deposition could depress *Sphagnum* C uptake up to 50-60 g C m^{-2} yr^{-1}. This is twice as large as recent estimates of annual C sequestration in northern peatlands based on long-term eddy covariance monitoring (Yu *et al.* 2011). Assuming 10% of the *Sphagnum* production eventually enters long-term storage in the anoxic zone (Rydin & Jeglum 2013), then long-term C sequestration of *Sphagnum* would be reduced by roughly 5 g C m^{-2} yr^{-1} in the most negatively affected peatlands. If this long term decrease in C sequestration by *Sphagnum* can be offset by increased production of vascular plant litter is highly uncertain as non-woody vascular plant litter is readily decomposed and is less likely to enter into the anoxic zone as peat (Rydin & Jeglum 2013). Moreover, reduced *Sphagnum* growth may decrease the self-regulation capacity and resilience of these ecosystems to further disturbances such as changes in climate or land-use.

Regions that would be affected most in the future are SW Sweden, Ireland, England, NW Germany, Netherlands and the Alps. Here the N deposition rates remain too high to halt negative N impacts, even in the best-case N deposition scenario without climate change. This suggests that the oceanic-temperate peatlands in Europe will continue to suffer under the predicted N deposition levels. The area estimated to be affected is likely underestimated in southern Europe because of the many small peatlands which may fulfil the CORINE size criteria. Many of these temperate peatlands are protected nature reserves and may continue to face lower *Sphagnum* production, invasion of vascular plants (Berendse *et al.* 2001; Tomassen *et al.* 2004) and continued losses of DON to groundwater and surface waters (Bragazza & Limpens 2004).

Prediction uncertainties

Despite the numerous N application studies carried out in peatlands, making quantitative estimates remains a challenge. In our study we did not include any uncertainties in climate change (e.g. Ruete *et al.* 2012) or N deposition scenarios. Instead we explored one model for climate change (A1) and N deposition (GEA-CLE) and compared them with a constant scenario, including all combinations. Sensitivity analyses showed that uncertainties in these variables did not much affect predictions. Likewise, uncertainty in peatland cover was not of major influence and point predictions were mostly influenced by model parameter uncertainties.

Predictions presented here are associated with considerable uncertainties, reflecting both parameter uncertainty, but also unknown variables affecting the effect of N deposition. It is reasonable to assume that a large proportion of the random variation among N application experiments is mostly associated with unknown microhabitat and species-specific responses and factors that could not be included here because of the limited amount of data. For example, different effects of N form (dry versus wet) and vascular plant cover. We argue that at the peatland scale our average effects are reasonable and realistic, although we should be cautious in our interpretation of the large effects reported (e.g. future *Sphagnum* production reduction by 50% in some regions). By using higher temperatures we are making predictions near the limits of model space. Nevertheless, these results suggest that interactions between the effect of N and climate need to be addressed in future research.

Another potential uncertainty resides in the assumption of our model that short term experiments (here 1-6 years) can be extrapolated over a longer time span. This assumption was supported by time series analyses in Limpens et al. (2011) which did not detect an effect of experiment length on the effect of N application. Accumulated N deposition (i.e. total N load) may, however, be as important as deposition rates since low deposition levels may over time reach critical loads. We were unable to predict N deposition effects for a large part of northern Europe, because N deposition change is too low to allow reliable modelling. N experiments in peatlands have so far focused on rather large N loads, preventing accurate assessment of the impact of a more realistic low, but chronic N deposition rates. Evidence from other ecosystems, such as forest, suggests that low but chronic N loads induced by experimental N fertilization of forests in Sweden, Finland and the USA for a period of about 10- 30 year causes enhanced carbon sequestration by increased tree growth without signs of N saturation (Högberg et al., 2006; Pregitzer et al. 2007; Hyvönen et al., 2008). However, at elevated N deposition, N saturation, occurrence of soil acidification and the elevated incidence of pest and diseases may lead to adverse impacts on forest growth (Magill *et al.* 2004). These results imply that experiments with high N loads in forests tend to overestimate adverse impacts as compared to low chronic loads. Our model suggest generally positive effects of low N levels on *Sphagnum* production, which is supported by the field observations in areas that received low N deposition (~ 0.5 g m^{-2} yr^{-1}) during an extensive time period (e.g. 50 years - Gunnarsson *et al*. 2000, 34 years - Vitt *et al*. 2003). While intact *Sphagnum*-dominated peatlands show a high resistance to disturbances such as moderate drainage or low-intensity fire, the disruption of keystone species such as *Sphagnum* may reduce the resilience of peatlands and cascade into disequilibrium or ecosystem change (Luo & Weng 2011). Nevertheless, in the case of N deposition, it is also possible that thresholds do not exist, or are below a detection limit, and the ecosystem is changing in a continuous manner (Payne *et al.* 2013).

Evaluating critical loads

Overall, our analyses support the critical load range of 0.5 to 1.0 g m^{-2} yr^{-1} for peatlands (Bobbink & Hettelingh 2011). The lower bound of that range suggests that negative N-deposition impacts already occur at a very small increase (0.2-0.3 g m^{-2} yr^{-1} assuming a pre-industrial N deposition of 0.2-0.3 g m^{-2} yr^{-1}) of N deposition. In some areas, *Sphagnum* production showed reductions at the minimum N deposition increase that could be reliably modelled in our study (0.4 g m^{-1} yr^{-1}), while in other areas a greater increase of N deposition (up to 1 g m^{-1} yr^{-1}) was needed to achieve a significant production decrease. Overall, our results suggest the current critical load holds for the range of climatic conditions predicted for 2050.

Conclusions

Our modelling analysis suggests that the negative impact of N deposition was rather limited throughout the 1900s, but the negative effects observed in the end of the 20th century (up to 25% production reduction) are likely to continue or even increase in the 21st century. Peatland-rich areas in Fennoscandia are, however, less impacted and *Sphagnum* production may even be temporarily stimulated in some areas. Our results stress the vulnerability of temperate peatlands to N deposition impacts and highlight the need for more studies on the effect of small but chronic elevations of N deposition. Furthermore, our results suggest the current critical loads for peatlands are also valid under predicted climate warming.

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Table 1. Results of the Hierarchical Bayes Linear Models (HBLM) with $\log_e$ response ratio. Negative coefficients indicate that an increase in the predictor depresses the response of *Sphagnum* to adding N. Categorical levels are compared to the intercept which is set to without P addition and without vascular plants. Upper and lower 95% credible intervals are given. Residual heterogeneity represents between study variation not explained by predictors. Of the total between study variation, 51% was explained by the included predictors. N=107.

	coefficients	lower-95%	upper-95%
Intercept	1.491	0.847	2.124
Nitrogen (N) application rate	-0.032	-0.056	-0.009
Background N deposition	-0.200	-0.393	-0.009
Mean July temperature	-0.049	-0.083	-0.014
Mean annual precipitation	-0.000372	-0.000741	-0.000013
Presence of vascular plants	-0.377	-0.585	-0.181
Phosphorus application	0.225	0.023	0.432
Residual heterogeneity (τ^2)	0.062	0.032	0.097

Table 2. Summary statistics of model predictions at the European scale (see Fig.1 for coverage). CLE N refers to the current legislation N emission scenario (GEA-CLE, here called CLE N) and A1 clim change to the A1 climate scenario. Constant N keeps N deposition at the mean of 2000-2010 and constant climate uses the mean of 1961-1990. Average effect of N deposition on *Sphagnum* production and *Sphagnum* C balance of *Sphagnum*-dominated peatlands are calculated as weighted means using peatland cover in grid cells as weights. Area indicates total peatland area where *Sphagnum* is estimated to have been, or to be, affected negatively or positively. The 1st and 99th percentiles are given to show the range of the grid cell predictions.

Period	Scenario	Average effect on production (1st, 99th %ile) (% change)	Average effect on C balance (1st, 99th %ile) ($g\ m^{-2}\ yr^{-1}$)	Area (mil. ha)	
				neg	pos
1900 - 1950		-	-	-	-
1950 - 2000		-6 (-46, 9)	-6 (-46, 9)	0.84	0.36
2000 - 2050	CLE N, A1 Clim Change	-12 (-65, 16)	-12 (-65, 16)	1.11	0.44
	CLE N, Constant climate	-5 (-58, 25)	-5 (-58, 25)	0.92	0.63
	Constant N, A1 Clim Change	-15 (-52, 0)	-15 (-52, 1)	1.54	0.05
	Constant N and Constant climate	-8 (-49, 11)	-8 (-49, 11)	1.22	0.37

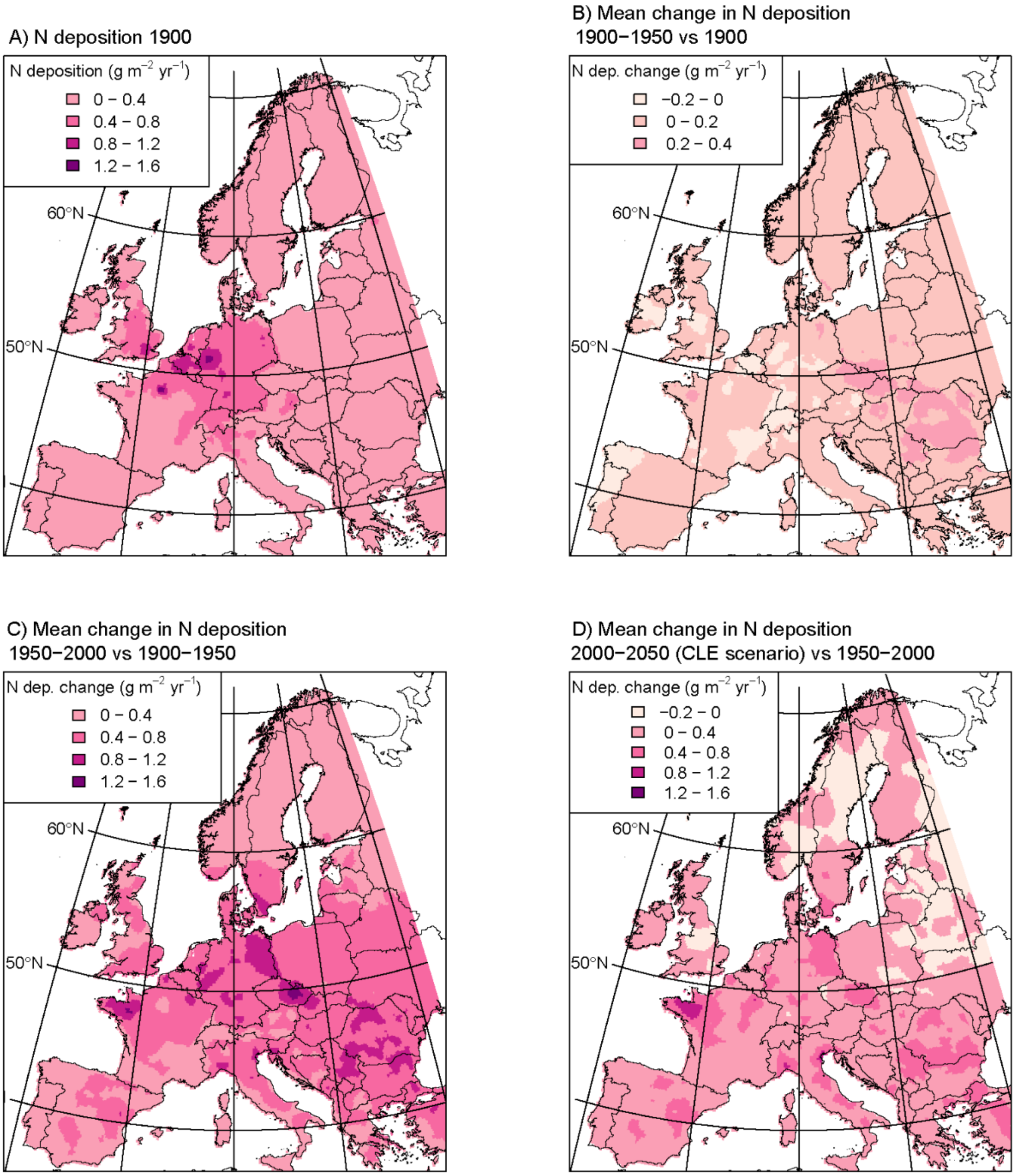

Fig 1. (A) Modelled nitrogen (N) deposition over Europe in the year 1900 and changes in N deposition over three time periods; (B) mean 1900-1950 compared to 1900, (C) mean 1950-2000 compared to the 1900-1950 mean, and (D), mean 2000-2050, compared to the 1950-2000 mean. Predictions for 2000-2050 (C) are based on the GEA-CLE deposition scenario.

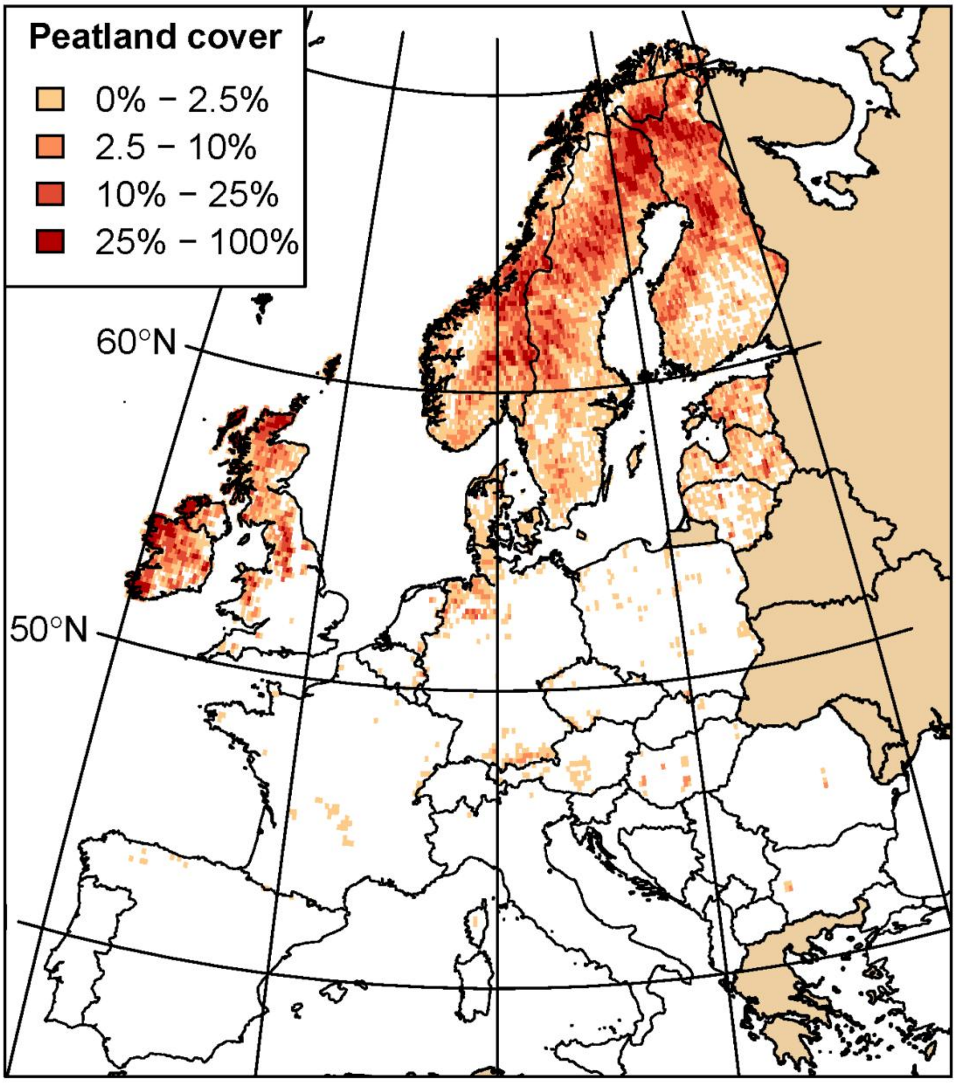

Fig. 2. Map showing the abundance (proportion of total land area) and distribution of open to semi-open *Sphagnum*-dominated peatlands in Europe. Data are based on the JRC forest map and the CORINE 2006 land cover map. The light brown colour indicates countries without data.

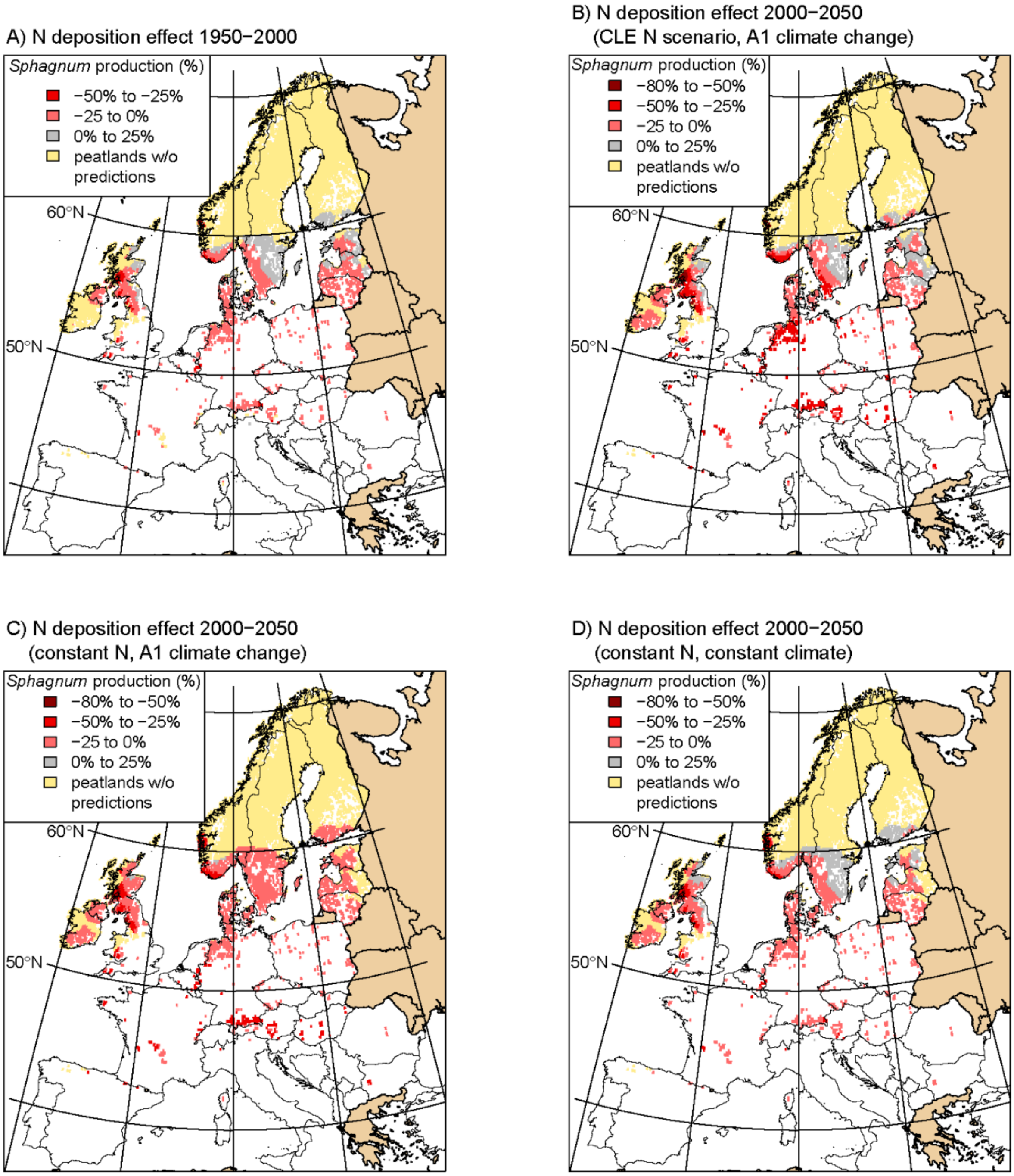

Fig 3. Effects of N deposition on *Sphagnum* production presented as percentage change relative to 1900. Annual mean effect between (a) 1950 to 2000, (b) 2000 to 2050 under a current legislation emission scenario (GEA-CLE, here called CLE N scenario) and the A1 climate change scenario, (c) 2000 to 2050 assuming constant N deposition (mean of 2000-2010) and climate change scenarioA1, (d) 2000 to 2050 assuming constant N deposition and constant climate (1961-1990 average). Only open to semi-open *Sphagnum* dominated peatlands were modelled. Yellow illustrates areas where N deposition were too low (< 0.4 g

m^{-2} yr^{-1} increase since 1900) to make reliable predictions. Light brown colour indicates countries without data on peatland distribution.

Supporting Information

Appendix S1. Figures illustrating the uncertainties of some of the predictions

Fig. S1. Prediction uncertainties - 90% credible intervals

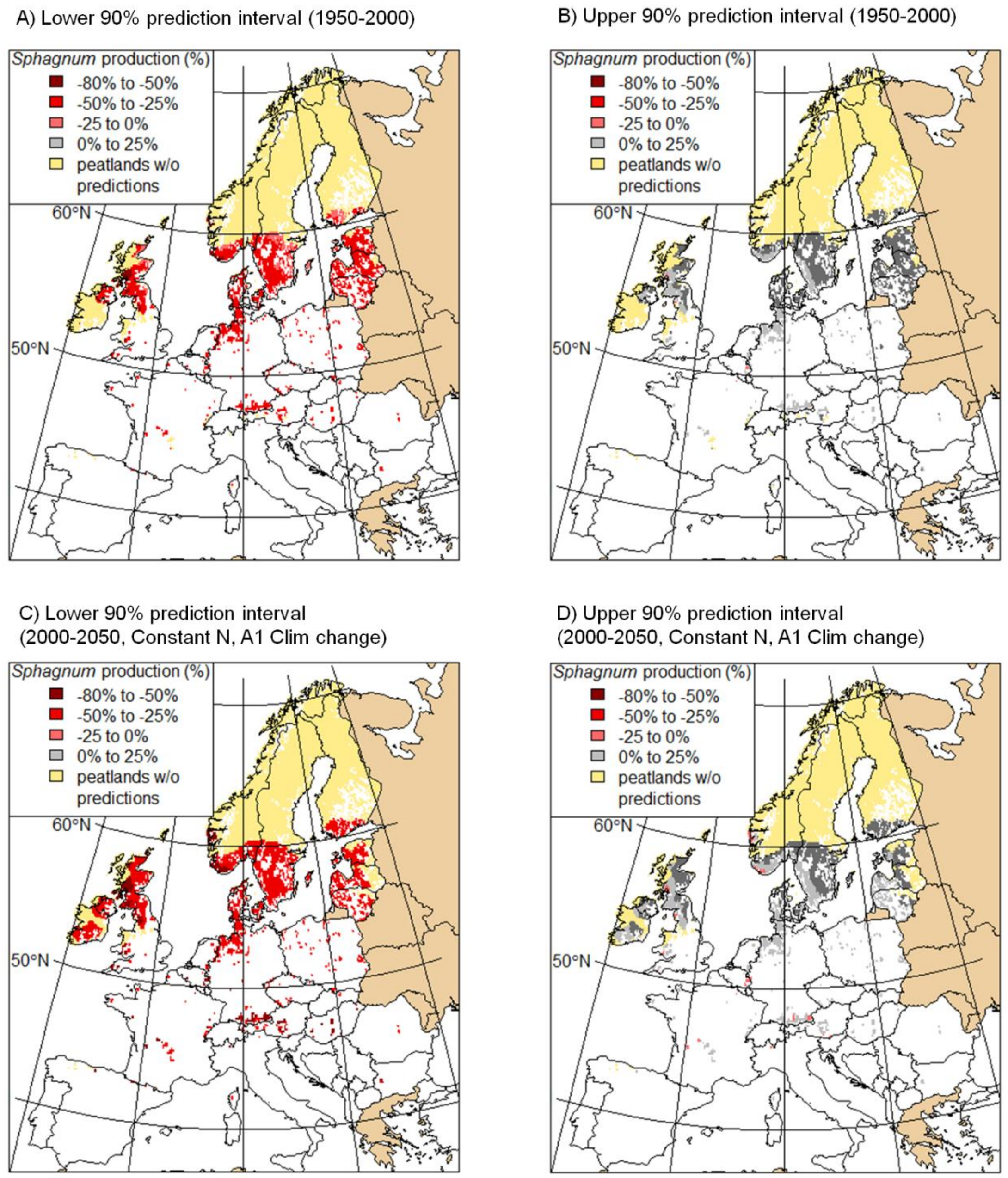

Fig. S1. Examples of prediction uncertainties (90% credible intervals) for two models, a-b) 1950-2000, and, c-d) 2000-2050 (Constant N, A1 Climate change scenario)

Fig. S2. Prediction uncertainties - 50% credible intervals

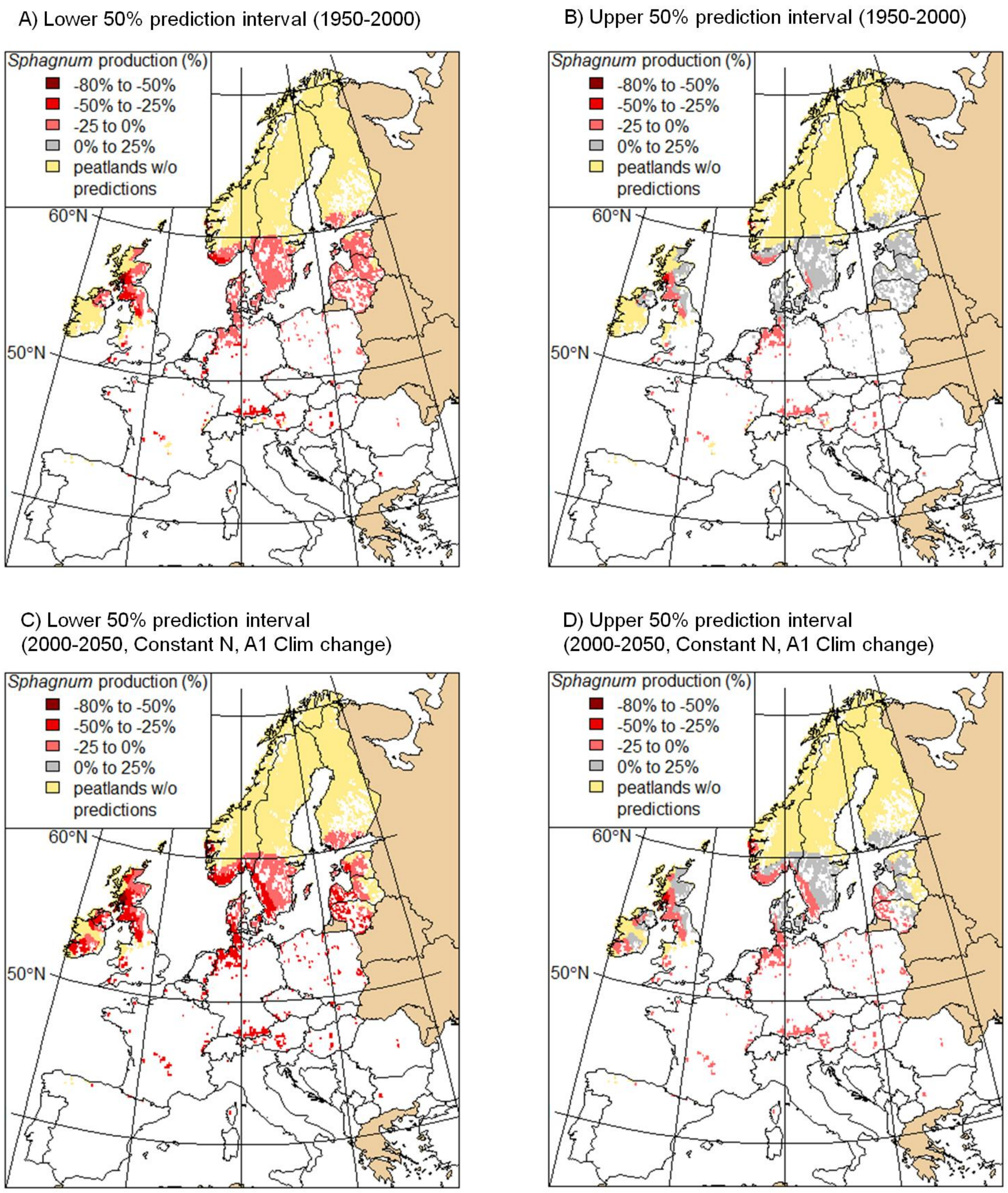

Fig. S2. Examples of prediction uncertainties (50% credible intervals) for two models, a-b) 1950-2000, and, c-d) 2000-2050 (Constant N, A1 Climate change scenario)