Increasing atmospheric CO₂ concentrations correlate with declining nutritional status of European forests

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The drivers of global change, including increases in atmospheric CO₂ concentrations, N and S deposition, and climate change, likely affect the nutritional status of forests. Here we show forest foliar concentrations of N, P, K, S and Mg decreased significantly in Europe by 5%, 11%, 8%, 6% and 7%, respectively during the last three decades. The decrease in nutritional status was especially large in Mediterranean and temperate forests. Increasing atmospheric CO₂ concentration was well correlated with the decreases in N, P, K, Mg, S concentrations and the increase of N:P ratio. Regional analyses indicated that increases in some foliar nutrient concentrations such as N, S and Ca in northern Europe occurred associated with increasingly favourable conditions of mean annual precipitation and temperature. Crucial changes in forest health, structure, functioning and services, including negative feedbacks on C capture can be expected if these trends are not reversed.
Atmospheric CO₂ concentrations and nitrogen (N) and sulfur (S) deposition, together with warming and drought, likely affect the nutritional status of forests1–6 and therefore their functioning, structure and ecosystem services7–9. Elevated atmospheric CO₂ concentrations, usually tested at 500–700 ppm, decrease the N and P concentrations of plants10–13. Increases in atmospheric CO₂ concentrations are frequently correlated with higher growth10 and more efficient photosynthesis, and thus likely dilute leaf-level nutrient concentrations. Increases in atmospheric CO₂ concentrations also reduce transpiration14 and stomatal conductance15, thus also hindering nutrient uptake16,17 that may even ultimately limit the initial increase in plant production under the rise of atmospheric CO₂ concentrations18–20. N deposition also increases tree productivity21,22 and foliar N concentrations but can decrease foliar P and Mg concentrations23–25. Warming tends to increase the mineralisation, cycling and availability of nutrients when water is available26 but the consequent increase in growth involves a dilution of nutrients that leads to frequent decreases in foliar N concentrations26,27 and increases in C:N ratios22,28. Plants at sites not limited by water respond by increasing nutrient uptake29,30, but if warming persists or even increases in the long term, nutrients can become limiting31,32. Warming in dry environments, though, can increase soil drought, exacerbating limitations of water and nutrients33,34. Plants under these conditions respond by activating mechanisms for conserving and taking up water and nutrients but C: nutrient ratios still frequently increase in photosynthetic tissues35–38.

The concentrations of atmospheric CO₂ have increased from ~350–360 ppm in the 1990s to the current 410 ppm (in 2019)39. The deposition of oxidised N in some regions of the world such as Europe peaked during the 1980s, up to 6–8-fold higher than in 1900, but has since decreased to half its highest value40,41. The annual deposition of reduced Nin Europe is currently more than two-fold higher than in 190042. S deposition in Europe has decreased to ~70% of the level in 190042. Europe has also warmed on species and foliar cohorts24,49 being found, with decreases, increases or no changes, depending in foliar P and Mg concentrations23–25. Warming tends to increase the mineralisation, cycling and availability of nutrients when water is available26 but the consequent increase in growth involves a dilution of nutrients that leads to frequent decreases in foliar N concentrations26,27 and increases in C:N ratios22,28. Plants at sites not limited by water respond by increasing nutrient uptake29,30, but if warming persists or even increases in the long term, nutrients can become limiting31,32. Warming in dry environments, though, can increase soil drought, exacerbating limitations of water and nutrients33,34. Plants under these conditions respond by activating mechanisms for conserving and taking up water and nutrients but C: nutrient ratios still frequently increase in photosynthetic tissues35–38.

These increasing CO₂ concentrations, changes in N and S deposition and climate change have been accompanied by a general decrease in foliar P concentrations and a consequent increase in N:P ratios in recent decades in Fagus sylvatica24,45–47, Picea abies and Pinus sylvestris24,48 and Quercus petraea24. Clear general patterns for foliar N concentrations, however, have not been found, with decreases, increases or no changes, depending on species and foliar cohorts24,49–52. Likely local, regional or latitudinal differences have not been considered, so these changes in foliar nutrient concentrations have not been consistently attributed to a particular environmental driver or combination of drivers. Furthermore, most reported nutritional changes in plants refer to N and P concentrations, but other important nutrients are key to the nutritional status of trees, such as K, S, Mg and Ca3.

We analysed (i) the changes in foliar elemental composition and stoichiometry during the last three decades for the main tree species in forests throughout Europe (Supplementary Fig. 1) at three different spatial scales: over the entire Europe, at different latitudes, and locally, as well as (ii) the empirical relationships of these changes with their possible drivers, i.e. increased atmospheric CO₂ concentrations, N and S deposition and climate change, using statistical attribution analyses, data available from field experiments and models of the responses of nutrients to these drivers of global change. The results showed that forest foliar concentrations of N, P, K, S and Mg decreased significantly in Europe by 5%, 11%, 8%, 6% and 7%, respectively, and that these decreases were especially large in Mediterranean and temperate forests and mainly related to the rising of atmospheric CO₂ concentration. In contrast, foliar N, S and Ca concentrations increased associated with increasingly favourable conditions of mean annual precipitation and temperature in boreal forests. Crucial changes in forest health, structure, functioning and services, including negative feedbacks on C capture can be expected if these trends in central and southern European forests are not reversed.

Results
Declining nutritional status. Foliar N, P and K concentrations decreased in European forests during the last three decades, by 5%, 11% and 8%, respectively (Fig. 1), especially in central and southern Europe (Fig. 2 and Supplementary Fig. 1). An exception is northern Europe where foliar N concentration increased and foliar P concentrations did not change. The foliar N:P ratio increased everywhere by an average of 7% (Fig. 1 and Supplementary Fig. 2). Foliar S and Mg concentrations decreased in Europe, by 6% and 7%, respectively (Fig. 3), although foliar S concentrations, as Ca concentrations, increased in northern Europe and decreased in central Europe (Fig. 4 and Supplementary Fig. 3). The trends were not dominated by any extreme values; the effect of the anomalously high or low years for Mg and S concentrations (Fig. 3) was minimised in the mixed model analyses. The results of the analyses after removing years 1996 and 2012 (respectively anomalously high and low) consistently showed that foliar Mg still decreased at a rate of −0.0036 ± 0.0007 mg g⁻¹ (P < 0.0001)—lower slope than in Fig. 3—and S decreased at a very similar rate as shown in Fig. 3 (−0.0030 ± 0.0007 mg g⁻¹, P < 0.0001).

The foliar elemental concentrations followed similar trends in all species (Fig. 2 and Supplementary Figs. 4–9), with few exceptions such as P and Mg increase in P. sylvestris at northern latitudes (Figs. 2 and 4). Each species had a distinct foliar elemental composition (elementome), even though the individual trees grew under different environmental conditions and limitations at distinct latitudes (see Fig. 5 and Supplementary Tables 3–10 for a DA and Supplementary Fig. 10 for similar results in a PCA; the DA and the PCA were applied to multi-elemental data space to quantify the ‘elementome’), result consistent with recent studies showing the strong species identity in foliar elemental composition33. But despite the observed species identity in their foliar elemental composition, all studied seven species, i.e. F. sylvatica, P. sylvestris, A. alba, P. abies, Q. ilex, Q. petraea and Q. robur, changed their elemental composition between 1990 and 2016. They shifted their foliar elemental composition along the axis toward decreasing foliar P, K and Mg concentrations and increasing N:P ratio during 2005–2016 relative to 1990–2004. The overall nutritional status of all species thus declined. The regional analysis, though, indicated that this decline did not extend to the northern forests. The foliar elemental composition of the trees at northern latitudes, mostly of P. sylvestris and P. abies, shifted toward increasing N but also to increasing N:P ratio (Fig. 5b) suggesting a softening of N limitation.

Possible drivers. CO₂ concentrations during 1990–2016 increased everywhere by ca. 50 ppm, N and S deposition decreased on average by ca. 25% and 65%, respectively, and temperature increased everywhere, especially in the north by almost 1 °C, whereas precipitation increased by ca. 50 mm year⁻¹ in the north and decreased by ca. 100 mm year⁻¹ in the south (Supplementary Fig. 11). The increase in atmospheric CO₂ was the only predictor systematically associated with the decreases in
nutrient concentrations (Fig. 6). The mixed-model analyses at the level of individual trees produced similar results (Supplementary Table 2). The regional analyses indicated that the increases in some foliar nutrient concentrations in northern Europe are not fully comparable with a progressive increase of atmospheric CO₂ concentrations in natural conditions where not only CO₂ concentrations change but also many other factors at the same time. For example, several European regions have become more arid in the last decades, especially in southern Europe, and rises of plant N:P ratios have been reported in response to increasing drought. Consistently, we have also observed that MAT has contributed significantly to increase foliar N:P ratios. FACE and OTC, instead, estimate the CO₂ effects by comparing treatment with control plots with all the other changes affecting equally to both treatment and control plots. In ICP data for European forests some other not controlled factors may have also contributed to decrease more foliar P than N concentration. The frequency and intensity of some forest pests have increased, so biotic factors not controlled in this study could have also favoured the P-uptake drop. Moreover, soil P availability tends to decrease through time by lower litter quality and the decreases in N and S deposition in recent decades. The experimental decrease may have been smaller also because the experiments test responses to increases in atmospheric CO₂ concentrations in a less sensitive range of higher CO₂ fluxes, method = "REML"). Statistically significant trends (in percentage of sites): N+: 5.68%, N−: 13.18%, P+: 1.21%, P−: 17.30%; N:P+: 14.72%, N:P−: 4.29%; K+: 2.02%, K−: 9.90%.

Discussion

The overall nutritional status of all species declined. The regional analysis, though, indicated that this decline did not extend to the northern forests. A general decline in plant nutritional status has also been reported in other regions such as North America.

Herbarium analyses also indicated that declines in plant N have already occurred in the last century despite increases in N deposition. Our findings are also consistent with a recent study suggesting a general global pattern of decreasing foliar N concentration ca. 9% over the last 40 years and with many examples of local to regional decreases in foliar N and P concentrations.

Consistently with our results, recent meta-analyses of elevated CO₂ experiments have found that rising CO₂ concentrations have led to decreases in N:P ratios in different plant tissues and woody plants. Deng et al. hypothesis that the experimental increases in atmospheric CO₂ concentrations stimulate higher plant uptakes of P than N. However, FACE and OTC experiments are not fully comparable with a progressive increase of atmospheric CO₂ concentrations in natural conditions where not only CO₂ concentrations change but also many other factors at the same time. For example, several European regions have become more arid in the last decades, especially in southern Europe, and rises of plant N:P ratios have been reported in response to increasing drought. Consistently, we have also observed that MAT has contributed significantly to increase foliar N:P ratios. FACE and OTC, instead, estimate the CO₂ effects by comparing treatment with control plots with all the other changes affecting equally to both treatment and control plots. In ICP data for European forests some other not controlled factors may have also contributed to decrease more foliar P than N concentration. The frequency and intensity of some forest pests have increased, so biotic factors not controlled in this study could have also favoured the P-uptake drop. Moreover, soil P availability tends to decrease through time by lower litter quality and the decreases in N and S deposition in recent decades. The experimental decrease may have been smaller also because the experiments test responses to increases in atmospheric CO₂ concentrations in a less sensitive range of higher CO₂ concentrations than the actual current range of 360–410 ppm.

The decrease in N deposition was slightly and positively related with foliar Mg and negatively with K concentrations. The decrease in S deposition was slightly and positively related with foliar K and negatively with Mg concentrations. The effects of the shifts in N and S deposition on foliar elemental composition during the studied period were thus weak. Sulfur deposition has dropped in general across Europe since 1980s, but N deposition despite having decreased in general at European scale in the last two decades, has not decreased in all sites, N loads, despite lower, continue being substantial, and, in general, no symptoms of...
Fig. 2 Geographical distribution of the annual rate of change for N, P, and K foliar concentration in mg g⁻¹. a the entire forests, b Pinus sylvestris, c Picea abies, and d Fagus sylvatica. The estimations are based on neural networks with MAP, MAT and nutrient deposition as inputs, using 80% of the trees with more than five measurements for training and 20% for validation. We replicated the process 1000 times and averaged the results. Map spatial resolution 0.1°.
significant recovery of soil status have been observed (Schmitz et al.\(^3\)), and the references therein). N deposition can cause deficiencies in other nutrients than N and nutrient imbalances due to a range of effects, including stimulation of plant growth (dilution effects) and negative effects on tree nutrient acquisition by modifying mycorrhizal associations\(^{24,66}\). Increasing mean annual temperature (MAT) also contributed to the decrease in Mg and the increase in Ca and Ni:P. Ca Moreover, in general, increases in soil pH translate into higher foliar cation concentrations\(^{67-69}\).

The higher temperatures at the northern European sites favoured longer growing seasons, biological activity and nutrient uptake, thus accounting for the lower general decreases in foliar nutrient concentrations and even the increase in N. In contrast, the increases in temperature and consequently in drought (decreases in mean annual precipitation (MAP) in southern Europe could account for the decline in soil fertility and capacity of nutrient uptake, all of which contribute to a decrease in foliar nutrient concentration, as in experimental drought studies conducted in Mediterranean forests where mineralisation, soil enzymatic activity and plant growth also decreased, thus leading to a large decrease in aboveground nutrient

*Fig. 3 Decreasing tree foliar Ca, Mg and S concentrations.* The black lines indicate the average trends, and the shaded areas indicate the standard errors of the average trends. The inset shows the percentages of forests with decreasing and increasing foliar nutrient concentrations. Red and blue indicate forests with increasing and decreasing trends, respectively. All values were adjusted to the same mean to remove forest-specific variability. See Supplementary Table 1 for detailed results of the model lme (foliar c variables = year, random = ||country/plot/species, data = daedas, method = "REML"). Statistically significant trends (in percentage of sites): Ca+: 7.46%, Ca−: 6.45%, Mg+: 7.11%, Mg−: 8.13%; S+: 6.09%, S−: 12.19%.

A widespread decline in crown condition, disruption of food webs and increased tree mortality with increased drought associated with climate change have also been reported for these southern European forests\(^{60}\).

Other foliar chemistry factors and processes not-measured in ICP Forest such as resorption or tree age could also be underlying the observed decrease of foliar nutritional condition. However, the leaves selected for foliar analyses are mature non-senescent leaves and the trees selected for foliar analyses are adult non-senescent trees, and given the frequent long life of trees, the changes in tree age during the studied period, ~25 years as maximum in the individual plots, should not affect much the foliar concentration. Furthermore\(^{35}\), reported that foliar N contents and dry weight tended to slightly decrease with age but that this was not the case for N concentrations in *Fagus sylvatica* and *Picea abies* in European forests.

Nutrient impoverishment can have multiple effects on the structure, function and ecosystem services of forests. For example, N and P are fundamental to C assimilation and protein synthesis, so their decreased concentrations could constrain the capacity to take up carbon and the effect of CO\(_2\) fertilisation in forests\(^8\). Foliar N:P ratios are negatively correlated with plant net photosynthesis and growth\(^3\), so the increasing foliar N:P ratios in European forests (Fig. 2) suggested a worsening nutrient imbalance that may partly account for a lower effect of CO\(_2\) fertilisation\(^8\). The consequent changes in plant C:N and C:P stoichiometries can also drive ecosystem-level N availability by the effects on litter quality decrease, microbial N immobilisation and mineralisation\(^{19}\). The reduction in the availability of N may in turn affect the efficiency of sequestration of C. Increases in foliar C and decreases in N and P are associated with increases in non-structural carbohydrates and carbon-based secondary metabolites\(^{41,74}\) and decreases in foliar protein content, thus decreasing the nutritional quality of plants\(^{65,75,76}\) for wildlife and livestock.

We thus conclude that foliar concentrations of N, P, K, S and Mg are generally decreasing in European forests. These decreases are generally larger for P than N, so the foliar N:P ratio has increased in most European forests. These trends are mostly associated with increasing atmospheric CO\(_2\) concentrations that have led to a higher nutrient demand by trees. The soil nutrient supply was probably not always sufficient to meet the growing demands by trees, which could partly explain the deterioration of tree mineral nutrition. These decreasing trends are stronger in southern and central Europe than in northern Europe where the concentrations of some elements are even increasing, all in consonance with the increase in MAT favouring nutrient availability and uptake in northern Europe while hindering them in increasingly dry southern Europe. These nutrient limitations for forest growth should be taken into account by the scientific and environmental management communities to avoid overestimations of forest productivity in response to elevated atmospheric CO\(_2\) when developing global climate models and projections. The consequences of such pervasive nutrient impoverishment can be key for forest structure, function, health and capacity to provide ecosystem services.

**Methods**

**Data acquisition**. We used foliar and growth data of the International Cooperative Programme on Assessment and Monitoring of Air Pollution Effects on Forests operating under the UNECE Convention on Long-range Transboundary Air Pollution (CLRTAP) (ICP Forests). Activities under ICP Forests are conducted in 42 states using harmonised sampling and analysis following the manuals on Sampling and Analysis of needles and leaves—Part X; http://www.icp-forests.org/pdf/manual/2016/ICP_Manual_2016_01_part12.pdf and Tree growth—Part V; (http://www.icp-forests.org/pdf/manual/2016/ICP_Manual_2016_01_part05.pdf).

In this study, we have gathered all the available data of foliar N, P, K, S, Ca and Mg...
Fig. 4 Geographical distribution of the trend in annual rate of variation for Ca, Mg, and S foliar concentration in mg g\(^{-1}\). a) the entire forests, b) *Pinus sylvestris*, c) *Picea abies*, and d) *Fagus sylvatica*. The estimations are based on neural networks using 80% of the trees with more than five measurements for training and 20% for validation. We replicated the process 1000 times and averaged the results.
concentrations in annual series. We have used data from 28 countries with a total of 528 different plots and with 506 of these plots with the canopy clearly dominated by one tree species, 21 co-dominated by two tree species, and 1 plot co-dominated by three tree species.

Foliar samples were taken at least biannually at the intensive forest monitoring plots (Level II) from the living crown of the dominant canopy layer providing information on the nutrient status of one or more species per plot. The analysis was done for 1000 needles or 100 leaves covering a range of elements (for detailed information please refer to the ICP Forest Manual). Briefly, in each plot a minimum of five dominant trees were randomly selected avoiding the trees used in crown assessment, so to avoid crown damage of these trees. A composite sample by mixtures (43%) followed by wet ashing (40%) and posterior determination, mostly by inductively coupled plasma coupled to atomic emission spectrometry (ICP-AES). Quality assurance and assessment of the analytical process was controlled by the regular organisation of Inter-laboratory Comparisons by the Forest Foliar Co-ordinating Centre. The laboratory results were considered of efficient quality when the laboratory receives a quality certificate for the concerning method.

Discriminant analyses of the foliar elemental concentrations and N:P ratios for the seven dominant species, Pinus sylvestris, Picea abies, Abies alba, Fagus sylvatica, Quercus robur, Quercus petraea and Quercus ilex, for the entire Europe and for northern, central and southern Europe. All plots compare the data for 1990–2004 with the data for 2005–2016. The circles/ellipses for each species and period depict the mean position and the space occupied by the 95% confidence interval for the scores of each species.

Statistics and reproducibility. First, we used time series of observations of foliar N, P, K, S, Ca and Mg concentrations at 410 European sites (Supplementary Fig. 12) for the last three decades (1990–2016) to investigate their temporal trends using mixed models, where year was the fixed covariate and site-species was the random factor. Second, we repeated the analyses separately for northern, central and southern European forests (separated by parallels at 46° and 58° North; Supplementary Fig. 1b). Third, we repeated the previous two analyses for each of the most abundant species: F. sylvatica, P. sylvestris, and P. abies. Fourth, we predicted the rate of changes in foliar elemental concentration across Europe as a function of MAP, MAT and nutrient deposition rate of change using neural networks. We calculated the rate of changes of the foliar elements for each tree with more than five measurements and the rate of change of the MAP, MAT and nutrient deposition for the same period and location using Theil-Sen robust regressions implemented in mblm R package71. Then, we used 80% of the estimated rates to train the neural networks and 20% for validation using keras78 in R79 with Tensorflow80 backend. The neural networks had two hidden layers with 128 units each with rectified linear activation functions. We repeated the process 1000 times making predicted maps and averaging the results. Finally we masked the pixels with no forest81 and for species specific models, we also applied a mask with the distribution maps82–84. Fifth, we used multivariate analyses, including a discriminant analysis (DA) and a principal component analysis (PCA) of all nutrient variables to analyse the shifts in the elementome53,85,86 for each species for the entire Europe and each of the three latitudes.

We explored which environmental factors could better explain the observed changes in foliar elemental concentrations. To do so, we estimated the temporal changes in foliar elemental concentrations. To do so, we estimated the temporal concentrations in annual series. We have used data from 28 countries with a total of 528 different plots and with 506 of these plots with the canopy clearly dominated by one tree species, 21 co-dominated by two tree species, and 1 plot co-dominated by three tree species.

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We next used the R metafor (v. 2.0-0) and forest plot (v. 1.7.2) packages to conduct a meta-analysis of published experimental data for the effects of elevated atmospheric CO₂ concentrations predicted by the final model (containing all significant predictors) and the trends of the foliar nutrient concentrations predicted by the same model but sequentially maintaining the temporal varying predictors constant (e.g. temporal anomalies in MAT were held constant using the median for each site, but all other predictors varied based on the observations). The differences between the predicted change in foliar nutrient concentrations were considered as unknown temporal contributions. All errors were calculated using error propagation.

We finally conducted a meta-analysis of published experimental data for the environmental factor that was best correlated with nutrient depletion: CO₂ fertilisation. We used the keywords: atmospheric CO₂, C:N, CO₂ fertilisation, C:P, decrease, dilution, FACE, foliar, increase, leaf, needle, nitrogen, N:P, phosphorus, photosynthetic tissues, rise, and stoichiometry in our web search in Web of Science and google scholar between 1988 and 2018. We gathered the available studies that tested for the effects of elevated atmospheric CO₂ concentrations (both using FACE and OTC methodologies) on N and P concentrations and the N:P ratio of green mature leaves for all types of vegetation (353, 297 and 684 studies, respectively). The list of the published articles considered appears in the supplementary Material. The R metafor (v. 2.0-0) and forest plot (v. 1.7.2) packages were used for the
analyses as described in Hedges et al.\textsuperscript{23}. We calculated the response ratio (InRR) as \( \ln(\text{Xt/ Xt}) = \ln(\text{Xt}) - \ln(\text{Xt}) \), where \( \text{Xt} \) and \( \text{Xt} \) are the values of each chemical compound in leaf tissue observation before and after the treatment respectively. The sampling variance for each InRR was calculated as \( \ln(\text{Xt/ Xt})^{2} \), where \( \text{Xt} \) and \( \text{Xt} \) are the post-treatment and control sample sizes, standard deviations, and mean response values, respectively. The sensitivity was evaluated per ppm of CO\(_2\) added in the treatment. We thereafter standardized the effect size to an increase of 50 ppm since atmospheric CO\(_2\) concentrations increased everywhere by ca. 50 ppm during 1990–2016.

**Reporting summary.** Further information on research design is available in the Nature Research Reporting Summary linked to this article.

**Data availability**

The datasets generated during and/or analysed during the current study are available from the corresponding author, Prof. Josep Penuelas.

**Code availability**

All codes are available upon request to the corresponding author, Professor Josep Penuelas.

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Competing interests
The authors declare no competing interests.

Additional information
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