**1.) Sampling locations, associated climatic conditions, and trends in leaf protein content.**

a.) Map of eastern Australia showing leaf sampling locations (triangle symbols).

Sampling locations were located along three latitudinal bands, spanning broad gradients of rainfall and temperature. A total of 324 photosynthetically active Eucalypt leaf samples were collected from 32 species; four species were recorded at multiple location. For each species-location combination, three canopy leaves were collected from each of three individuals to make a total of nine samples;

b.) Mean annual temperature (oC) and mean annual precipitation (mm, log scaled) at sampling sites, derived from the Australian Bureau of Meterology’s Australian Water Availability Project (time series spanning 1950-2015);

leaf protein content per unit leaf area declines with increasing temperature (c, R2 = , % decline = ) and rainfall (d, R2 = , % decline = ). Each point shown represents an individual leaf sample, vertical bands of points represent data for multiple species in a given climate setting.

e.) Leaf protein content is closely linked to leaf nitrogen content (R2 = , % increase = ).

**2.) Linking leaf protein abundances with environment and functional traits.**

**2a.)** Protein functional composition of eucalypt leaves. We used a hierarchical protein functional annotation system (MAPMAN/Mercator, ref) to assign proteins to functional groupings. Here we show the average abundances of proteins associated with the major functional groupings in eucalypt leaves; angular fraction indicates the proportion of protein associated with a named functional category. The hierarchical annotation scheme is represented by the layers of the plot: the innermost layer corresponds to the broadest categories in the scheme, e.g. ‘photosynthesis’. Moving outwards, protein amounts are annotated to progressively more specific functions, e.g. ‘light harvesting complex II’. The majority of protein in leaves is associated with photosynthesis (green); protein synthesis, folding and degradation is the second largest top-level category (blue).

**2b.)** We can now directly investigate relationships between environmental variables, leaf traits and the abundance of proteins associated with functions of interest. Pearson correlations between pairs of variables are represented by coloured tiles where p < 0.05. Relative protein abundances (i.e. as a proportion of total leaf protein abundance) are used to calculate correlations presented in the top/left diagonal and protein abundances on a per leaf area basis (mg protein / m2 leaf area) are used to calculate correlations presented in the bottom/right diagonal.

3.) **Trends in abundance of light harvesting and carbon assimilation proteins across continental-scale environmental gradients.**

Trends in abundance of photosystem proteins [symb, col], rubisco [symb, col], and other Calvin cycle enzymes [symb, col] across gradients of: a,b.) canopy-corrected mean annual irradiance (MJ/m2/year), R2 = , modelled change (%) = , p = ; c,d.) mean annual precipitation (mm/year) R2 = , modelled change (%) = , p = ; e,f.) mean annual temperature R2 = , modelled change (%) = , p = . A full table of univariate regression statistics is provided in the supplementary materials.

Points represent the average protein abundance for individual species \* site combinations (n = 9, 3 leaves from each of 3 individuals). SEM error bars are presented for protein abundances and for canopy-corrected irradiances, since mean canopy openness values are derived from measurements of three individuals. Model fits (OLS regression) are shown where p < 0.05.

The top row of the panel (2 a,c,e) shows models fit using protein abundances expressed on a proportional basis (i.e. as a proportion of total leaf protein abundance); the bottom row shows models fit using protein abundances expressed on a per leaf area basis (mg protein / m2 leaf area).

4.) **Abundance of photosynthetic proteins in relation to total leaf protein content and leaf mass per area.**

Trends in abundance of photosystem proteins [symb, col], rubisco [symb, col], and other Calvin cycle enzymes [symb, col] in relation to: a,b.) leaf mass per area (g/m2), R2 = , modelled change (%) = , p = ; c,d.) total leaf protein (mm/year) R2 = , modelled change (%) = , p = . A full table of univariate regression statistics is provided in the supplementary materials.

Points represent the average protein abundance for individual species \* site combinations (n = 9, 3 leaves from each of 3 individuals). SEM error bars are presented for protein abundances and for canopy-corrected irradiances, since mean canopy openness values are derived from measurements of three individuals. Model fits (OLS regression) are shown where p < 0.05.

The top row of the panel (2 a,c,e) shows models fit using protein abundances expressed on a proportional basis (i.e. as a proportion of total leaf protein abundance); the bottom row shows models fit using protein abundances expressed on a per leaf area basis (mg protein / m2 leaf area).