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Intramolecular stable isotope distributions detect plant metabolic responses on century time scales

Jürgen Schleucher (1), Ina Ehlers (1), Angela Augusti (2), and Tatiana Betson (1)

(1) Medical Biochemistry&Biophysics, Umeå University, S-90187 Umeå, Sweden (jurgen.schleucher@chem.umu.se), (2) Institute of Agro-environmental and Forest Biology, NCR, 2 0510 Porano (TR), Italy

Plants respond to environmental changes on a vast range of time scales, and plant gas exchanges constitute important feedback mechanisms in the global C cycle. Responses on time scales of decades to centuries are most important for climate models, for prediction of crop productivity, and for adaptation to climate change. Unfortunately, responses on these timescale are least understood.

We argue that the knowledge gap on intermediate time scales is due to a lack of adequate methods that can bridge between short-term manipulative experiments (e.g. FACE) and paleo research. Manipulative experiments in plant ecophysiology give information on metabolism on time scales up to years. However, this information cannot be linked to results from retrospective studies in paleo research, because little metabolic information can be derived from paleo archives.

Stable isotopes are prominent tools in plant ecophysiology, biogeochemistry and in paleo research, but in all applications to date, isotope ratios of whole molecules are measured. However, it is well established that stable isotope abundance varies among intramolecular groups of biochemical metabolites, that is each so-called "isotopomer" has a distinct abundance. This intramolecular variation carries information on metabolic regulation, which can even be traced to individual enzymes (Schleucher et al., Plant, Cell Environ 1999).

Here, we apply intramolecular isotope distributions to study the metabolic response of plants to increasing atmospheric [CO_2] during the past century. Greenhouse experiments show that the deuterium abundance among the two positions in the $C6H_2$ group of photosynthetic glucose depends on [CO_2] during growth. This is observed for all plants using C_3 photosynthesis, and reflects the metabolic flux ratio between photorespiration and photosynthesis. Photorespiration is a major C flux that limits assimilation in C_3 plants, which encompass the overwhelming fraction of terrestrial photosynthesis and the vast majority of crop species. To access century time scales, we traced this metabolic signal in historic material of two crop species during the past 100 years and find the same response as predicted from the greenhouse experiments. This allows estimating how much photorespiration has been reduced due to the anthropogenic CO_2 emission during the 20^{th} century, and shows that plants have not acclimated to increasing [CO_2] during more than 100 generations.

In summary, we demonstrate that metabolic responses of plants to environmental changes create intramolecular isotope signals. These signals can be identified in manipulation experiments and can be retrieved from plant archives. The isotope abundance of each intramolecular position is set by specific isotope fractionations, such as enzyme isotope effects or hydrogen exchange with xylem water (Augusti et al., Chem. Geol. 2008). Therefore it may be possible to simultaneously reconstruct several physiologic or climate signals from an archive of a single molecule.

The principles governing intramolecular isotope distributions are general for all metabolites and isotopes (D, ^{13}C), therefore intramolecular isotope distributions can multiply the information content of paleo archives. In particular, they allow extraction of metabolic information on long time scales, thereby connecting plant physiology with paleo research.