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CO_2-induced terrestrial climate feedback mechanism: From carbon sink to aerosol source and back

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Feedbacks mechanisms are essential components of our climate system, as they either increase or decrease changes in climate-related quantities in the presence of external forcings. In this work, we provide the first quantitative estimate regarding the terrestrial climate feedback loop connecting the increasing atmospheric carbon dioxide concentration, changes in gross primary production (GPP) associated with the carbon uptake, organic aerosol formation in the atmosphere, and transfer of both diffuse and global radiation. Our approach was to combine process-level understanding with comprehensive, long-term field measurement data set collected from a boreal forest site in southern Finland. Our best estimate of the gain in GPP resulting from the feedback is 1.3 (range 1.02–1.5), which is larger than the gains of the few atmospheric chemistry-climate feedbacks estimated using large-scale models. Our analysis demonstrates the power of using comprehensive field measure-

ments in investigating the complicated couplings between the biosphere and atmosphere on one hand, and the need for complementary approaches relying on the combination of field data, satellite observations model simulations on the other hand.

Introduction

Anthropogenic emissions of greenhouse gases increased substantially during the past century, being the most important forcing agents responsible for global warming (IPCC 2013). However,

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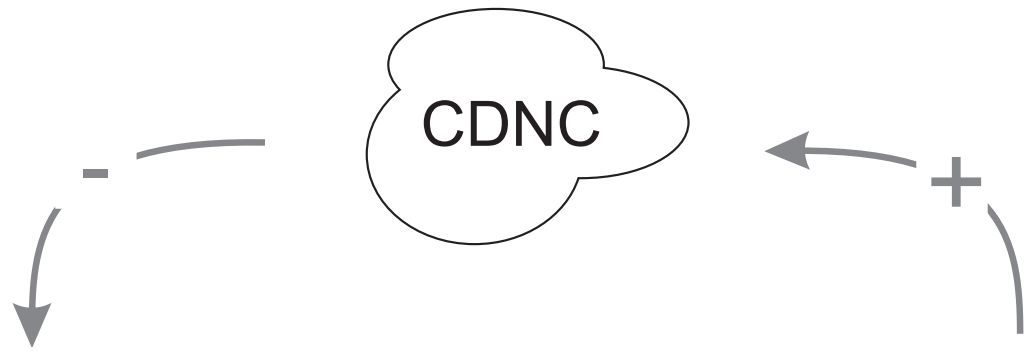
it is not straightforward to attribute or predict the climate change in detail because the internal variability of climate is only partially understood. One of the main reasons is the uncertainty associated with radiative forcing of aerosols and aerosol–cloud interactions, and that the climate systems includes a number of feedback mechanisms amplifying or dampening the original forcing, parameters that are difficult to quantify.

The continental biosphere plays an important role in the climate system by affecting the accumulation of carbon dioxide and other greenhouse gases in the atmosphere (Heimann and Reichstein 2008, Ballantyne *et al*. 2012), and by acting as a major source of natural aerosol parti-

cles and their precursors (Pöschl 2005, Guenther *et al*. 2012). Kulmala *et al*. (2004) suggested a negative climate feedback mechanism whereby higher temperatures and CO_2-levels boost continental biomass production, leading to increased biogenic secondary organic aerosol (BSOA) and cloud condensation nuclei (CCN) concentrations, tending to cause cooling in a manner similar to the CLAW hypothesis that linked climate change with the ocean biochemistry (Charlson *et al*. 1987, Quinn and Bates 2011).

Kulmala *et al*. (2013) extended the idea of the continental biosphere-aerosol-cloud-climate (COBACC) feedback mechanism further by adding the connection between aerosol particles, radiation and gross primary production (GPP) which is a measure of ecosystem-scale photosynthesis. As a result, the COBACC feedback mechanism has two major overlapping feedback loops, both initiated by increasing CO_2 concentrations and acting toward suppressing global warming (Fig. 1). An additional direct effect

might operate via changing CO_2 concentrations affecting BVOC emissions directly, which has been found in a number of experiments focused on isoprene emissions (for review of studies, *see* Arneth *et al*. 2011). It is unknown yet, whether a similar inhibitory effect can be found for monoterpene emissions. The focal points of the two loops are the ambient temperature and GPP, tied closely with aerosol–cloud interactions and terrestrial carbon sink, respectively. It is important to point out that many of the quantities and processes related to these loops are affected by human activities, and that there



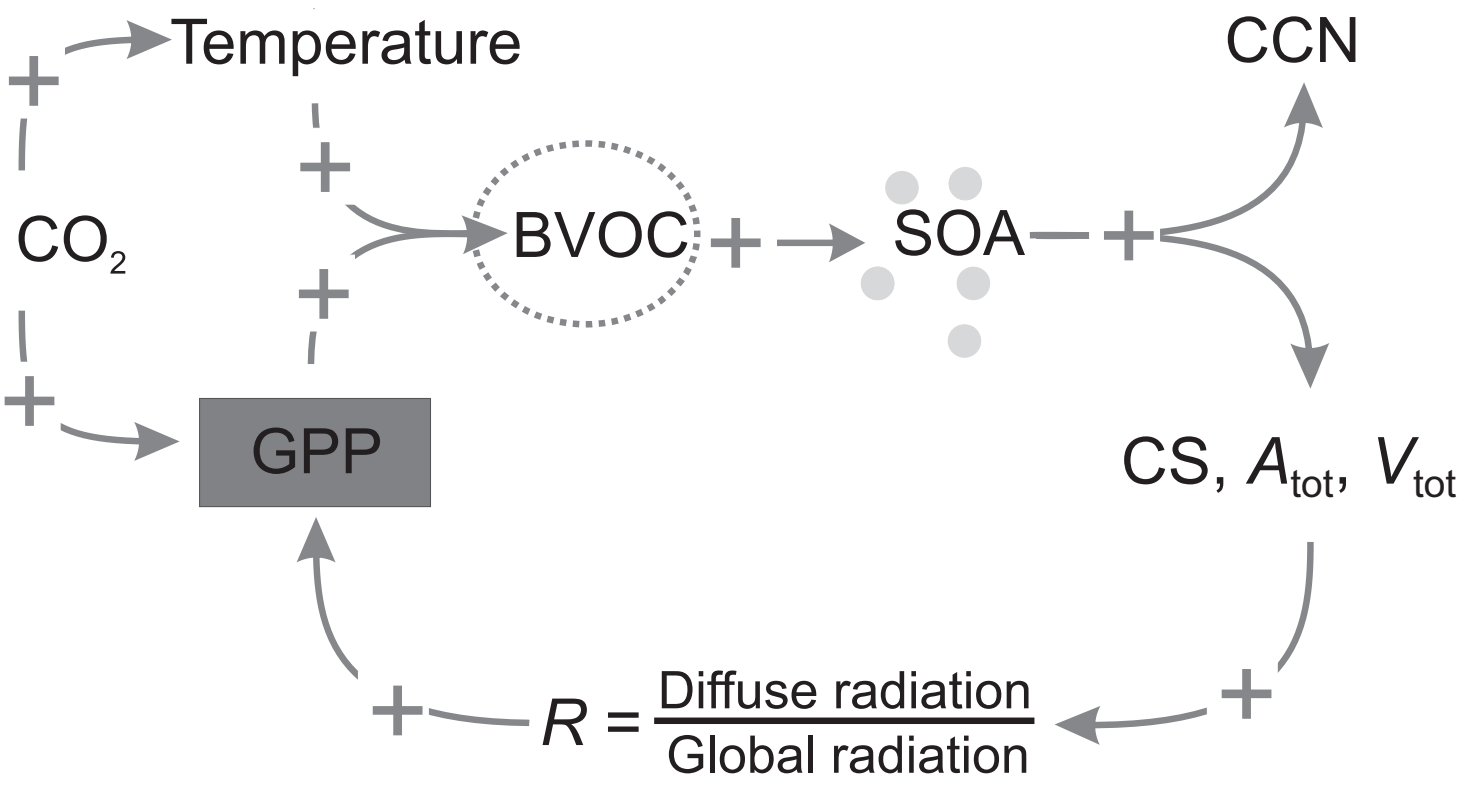


Fig. 1. The two feedback loops associated with the COBACC feedback. Here GPP is the gross-primary production, CS is the condensation sink, A_{tot} and V_{tot} are the total aerosol surface area and volume concentrations, respectively, and R is the ratio between diffuse and global radiations. BVOC refers to the biogenic volatile organic compounds, SOA to the secondary organic aerosol, GR_{org} to the particle growth rate caused by the compounds resulting form BVOC oxidation, CCN to the cloud condensation nuclei, and CDNC to the cloud droplet number concentration.

are many other feedback mechanisms that affect

some sub-group of the relevant quantities. With that in mind, the COBACC feedback can be considered a broad framework, which connects the human activities, the continental biosphere, and the changing climate conditions (*see* also Arneth *et al*. 2010).

The individual steps of the upper branch of the COBACC feedback mechanism have been investigated actively during the recent years, and strong support for the existence of this branch of the feedback has been obtained (e.g. Carslaw *et al*. 2010, Kerminen *et al*. 2012, Makkonen *et al*. 2012b, Paasonen *et al*. 2013, Rap *et al*. 2013). However, no systematic study on the lower branch of the COBACC feedback mechanism has been conducted so far. Here, we provide the first quantitative estimate on the strength of the lower-branch feedback loop using 15 years of continuous measurement data from a boreal forest site in Finland. We outline our general approach, describe the measurement data used in our analysis, estimate the strength of the

feedback loop and the associated uncertainties, and finally discuss the needs for future work.

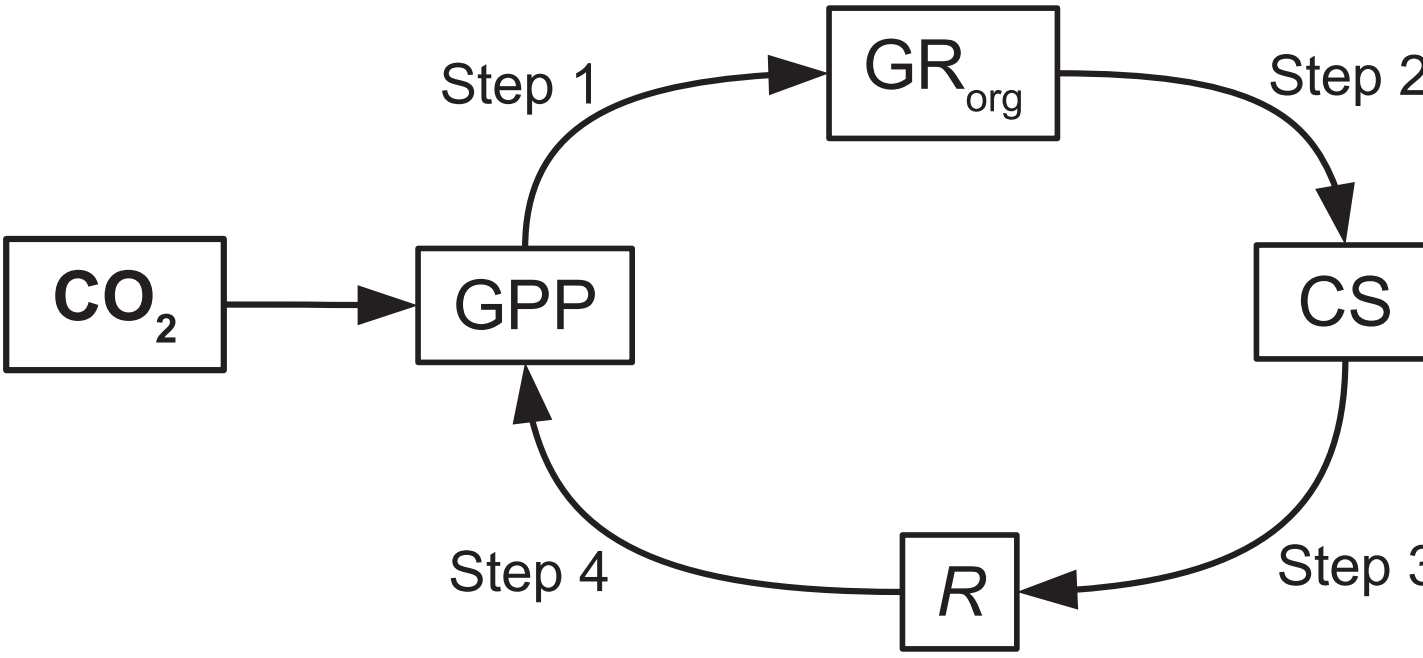


Fig. 2. The part of the COBACC feedback loop investigated here and the four steps in it investigated separately. Here GPP is the gross-primary production, GR_{org} refers to the particle growth rate caused by the compounds resulting from the oxidation of biogenic volatile organic compounds, and *R* is the ratio between diffuse and global radiation.

Material and methods

General approach

In order to determine the overall strength of the lower feedback loop in Fig. 1, we first divided the loop into four subsequent steps by selecting four key quantities inside the loop (Fig. 2), and then estimated how much a given change in any of these quantities changes the following quantity along the loop. The changes in key quantities, i.e. the strengths of the individual steps of the feedback loop, were estimated from long-term measurement data.

Our choices for the quantities to be looked at inside the loop were the forest gross primary production (GPP), particle growth rate due to organic vapour condensation (GR_{org}), condensation sink (CS), and the ratio between the diffuse and global radiation (R). We selected GR_{org} as a measure of the strength of atmospheric BVOC oxidation because this quantity is a good proxy for the gas-phase concentration of low-volatile

vapours resulting from atmospheric BVOC oxidation (e.g. Kulmala *et al*. 1998, Paasonen *et al*. 2010). As a measure of the amount of biogenic secondary organic aerosol, we selected the condensation sink, CS. This quantity describes the ability of the pre-existing aerosol particle population to remove condensable vapor molecules from air, in addition to which it is a relatively good proxy to the aerosol light scattering coefficient (Virkkula *et al*. 2011). The ratio between the diffuse and global radiation ties the aerosol load with GPP as this ratio tends to increase with an increasing aerosol load (e.g. Anton *et al*. 2012), and because higher values of *R* tend to enhance photosynthesis and GPP (Mercado *et al*. 2009).

After selecting the quantities, the four steps of the feedback loop are now that between GPP and GR_{org} (Step 1), that between GR_{org} and CS (Step 2), that between CS and *R* (Step 3), and that between *R* and GPP (Step 4) which closes the loop (Fig. 2). Step 0, i.e. the change in GPP

due to increasing atmospheric CO_2 concentrations, does not affect the overall strength of the feedback loop, yet such a change needs to exist to motivate our investigation. By comparing the values of GPP averaged over May–August each year at our measurement site with the corresponding values of the atmospheric CO_2 mixing ratio measured at the Global Atmosphere Watch station at Mauna Loa, Hawaii, Kulmala *et al*. (2013) demonstrated a clear, positive correlation between the values of these two quantities. The CO_2 mixing ratio increased almost 30 ppm over the time period of our investigation (1996–2011) both globally and at Mauna Loa (http://www.esrl.noaa.gov/gmd/ccgg/trends). At our measurement site, GPP increased 14% over the period 1996–2011 and the increase in the CO_2 mixing ratio since 2006 has been somewhat larger than that observed either globally or at Mauna Loa (Keronen *et al*. 2014). In the analysis presented later, we assume a 10% increase in GPP and scale changes in other quantities against that

value.

Data and analyses

The measurement data used in our analyses were obtained between 1996 and 2011 at the SMEAR II station (61°51´N, 24°17´E, 181 meters a.s.l.) in Hyytiälä, southern Finland. At SMEAR II, comprehensive measurements of exchange processes between the atmosphere and land ecosystem are being performed continuously (Hari and Kulmala 2005). The station and its surroundings represent a typical boreal coniferous forest dominated by Scots pine (*Pinus sylvestris*).

The values of GPP were calculated with a one-day time resolution as the difference

between the total ecosystem respiration (TER) and net ecosystem exchange (NEE) of CO_2:

$$GPP = TER - NEE. \quad (1)$$

Here, NEE was measured directly with the eddy covariance technique (Markkanen *et al*. 2001, Suni *et al*. 2003), whereas the values of TER were taken from model calculations based on nighttime NEE measurements (Suni *et al*. 2003, Kulmala *et al*. 2004). The GPP data were available from the years 1997–2011.

The growth rate GR_{org} was determined as the difference between the measured particle growth rate, GR, and growth rate due to sulphuric acid condensation, GR_{SA}.

$$GR_{org} = GR - GR_{SA}. \quad (2)$$

The value of GR used in our analysis was the average nucleation mode particle (3–25 nm) growth rate between the hours of 09:00 and 15:00. This restricted the GR data for the days when a clear nucleation event followed by new

particle growth to larger sizes was observed (e.g. Kulmala *et al*. 2012). The values of GR were calculated from the aerosol number size distribution data measured with a differential mobility particle sizer (DMPS) using the methods described by Kulmala *et al*. (2012). The value GR_{SA} were calculated using the method by Nieminen *et al*. (2010) assuming a gaseous sulphuric acid concentration that was taken from the proxy derived by Petäjä *et al*. (2009) using the measured SO_2 and global radiation as inputs.

The condensation sink, CS, was calculated from the aerosol number size distributions measured with a 10-min time resolution using DMPS according to (e.g. Kulmala *et al*. 2012):

$$\mathrm{CS} = 2\pi D \sum_{\mathrm{i}} \beta_{\mathrm{m}} d_{\mathrm{p},i} N_i. \qquad (3)$$

Here, D is the vapor diffusion coefficient of the condensing vapor, typically assumed to be sulphuric acid, $d_{\mathrm{p},i}$ is the diameter of particles in the size bin i, N_i is their number concentration, and

β_m is the Fuchs-Sutugin correction factor that takes into account the non-continuum regime effects for vapour condensation onto aerosol particles. Both CS and GR data were available for the whole period considered in our analysis (1996–2011).

The diffuse and global radiation was measured with a pyranometer (Reemann TP 3 and Middleton Solar SK08). The radiation fluxes, and hence the ratio R, were obtained with a 3-min time resolution. The necessary radiation data were available from the years 2000–2010.

For each year, we selected the period 1 March to 31 August for our analysis, which roughly corresponds to the biologically active part of the year. To minimize the influence of clouds on our analysis, especially on radiation fluxes, we considered cloud-free conditions only. Such conditions were determined using the brightness parameter, P, which is the daily ratio of the summed global radiation to the theoretical radiation sum, i.e. the maximum amount of

solar radiation that can be received in totally cloud-free conditions (*see* Kulmala *et al*. 2010). As threshold values for cloud-free and cloudy days we used $P > 0.6$ and $P < 0.3$, respectively, derived from comparisons of the brightness parameter to the cloudiness estimated from satellite images (Sogacheva *et al*. 2008). Since BVOC emissions depend not only on GPP but also on temperature, Steps 1 and 4 involving GPP were investigated by dividing the measurement data into 5-K temperature bins, and by looking at each temperature bin separately. In case of Step 4, the data were further divided into 100 W m^{-2} global radiation bins.

The strength of Steps 1–4 of the feedback loop were determined by fitting a straight line to the data points of each individual step using the bivariate fitting method described in more detail by Cantrell (2008).

Estimating the strength of the feedback loop

When looking at the individual steps of the considered feedback loop (Figs. 3–6), we found a moderate positive correlations between GPP and GR_{org} (Step 1), between GR_{org} and CS (Step 2), and between R and GPP (Step 4). The correlation was weakest, yet also positive, between CS and R (Step 3) due the highest scatter in data points between these two quantities. The esti-

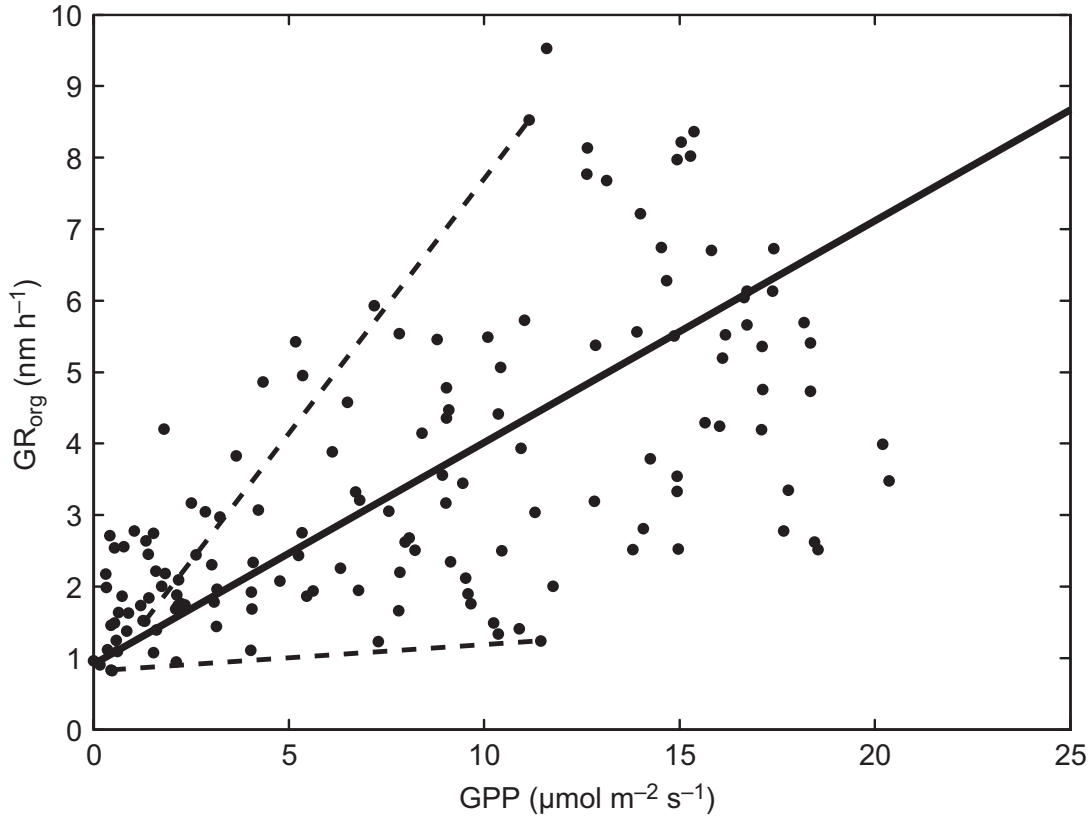


Fig. 3. The particle growth rate caused by the compounds resulting form BVOC oxidation, GR_{org}, obtained during nucleation event days as a function GPP averaged over the time period 09:00–15:00 during the same days. Only the days with the average temperature in the range 18–23 °C during 09:00–15:00 were taken into account. The solid line shows the least-squares fit to the measurement points (slope = 0.31 nm h^{-1} μmol^{-1} m^2 s, $r = 0.63$, $n = 148$, $p = 2.5 \times 10^{-17}$), and the dashed lines indicate the range of slopes used for upper- and lower-limit estimates. The data covers the years 1997–2011.

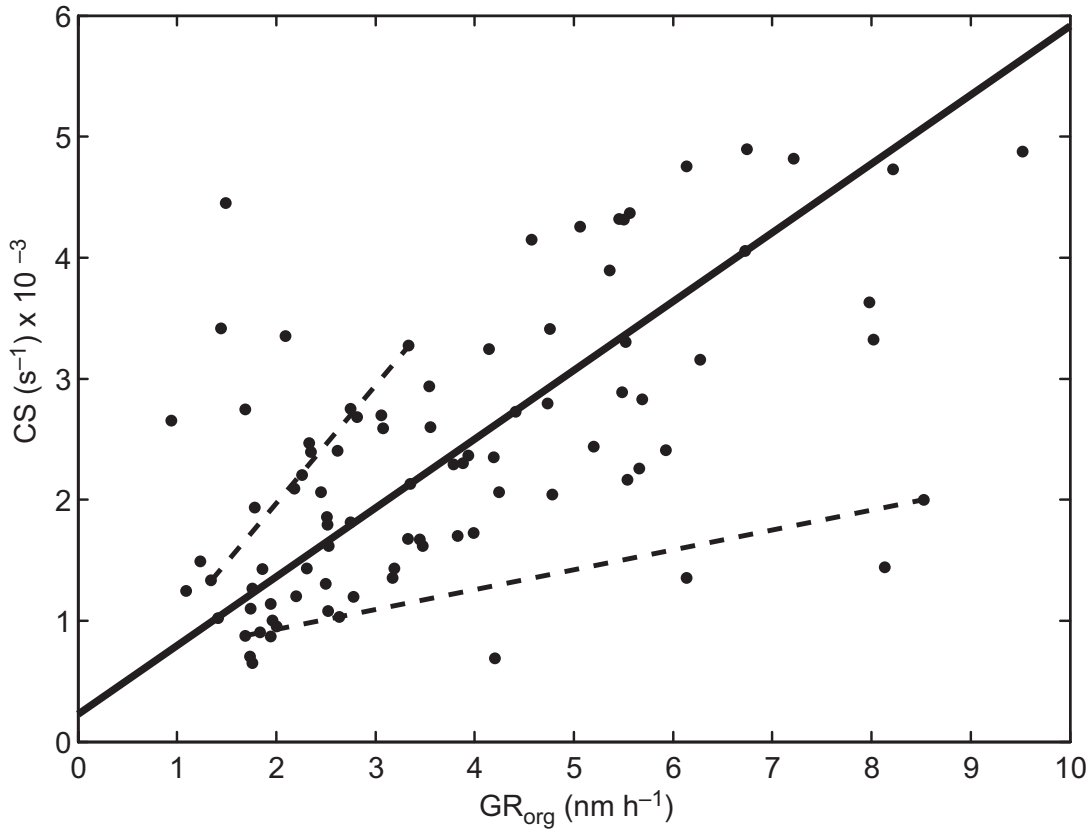


Fig. 4. The condensation sink, CS, as a function of the particle growth rate caused by the compounds resulting form BVOC oxidation, GR_{org}, in those nucleation event days when the measured air masses originated from the "clean" sector. The values of CS represent an average over the time period 15:00–23:00, which is when biogenic SOA formation is most evident in the measurement data. The solid line shows the least-squares fit to the measurement points (slope = 5.7×10^{-4} s^{-1} nm^{-1} h, $r = 0.60$, $n = 92$, $p = 9.3 \times 10^{-10}$), and the dashed lines indicate the range of slopes used for upper- and lower-limit estimates. The data covers the years 1996–2011.

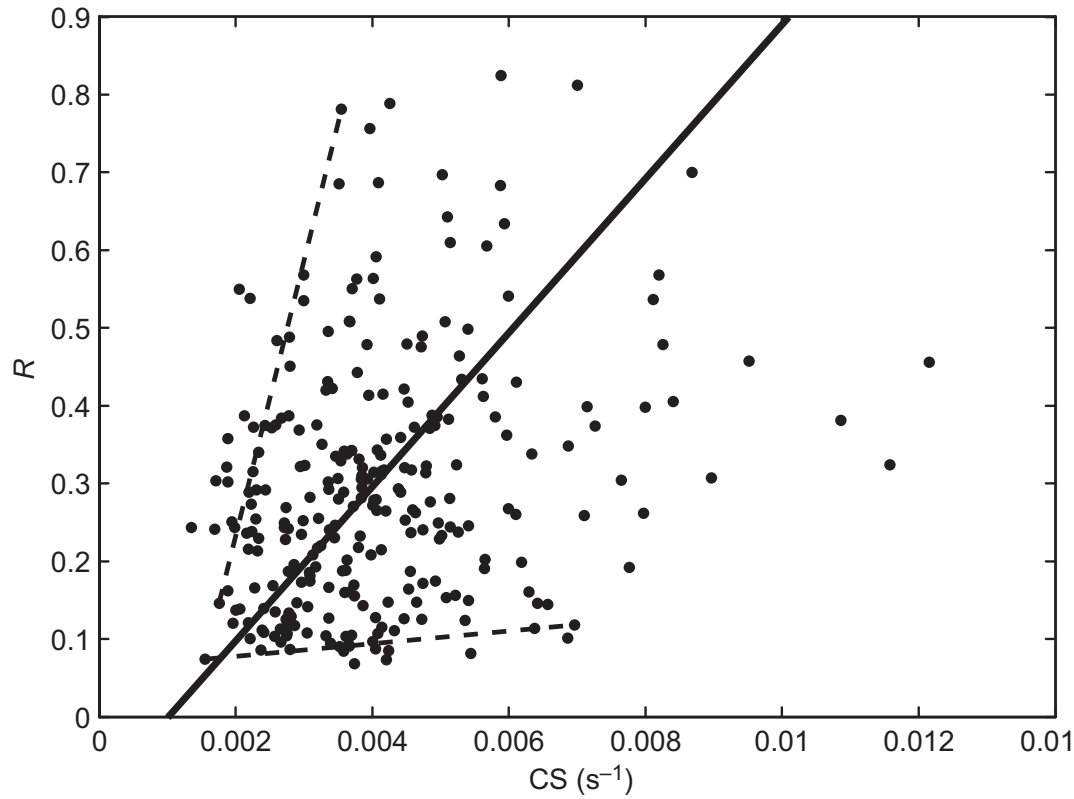


Fig. 5. The ratio between diffuse and global radiation, *R*, as a function of the condensation sink, CS, averaged over the time period 08:00–20:00 in days when the corresponding average temperature is in the range 18–23 °C. The solid line shows the least-squares fit to the measurement points (slope = 99 s, $r = 0.27$, $n = 446$, $p = 7.3 \times 10^{-6}$), and the dashed lines indicate the range of slopes used for upper- and lower-limit estimates. The data covers the years 2000–2010.

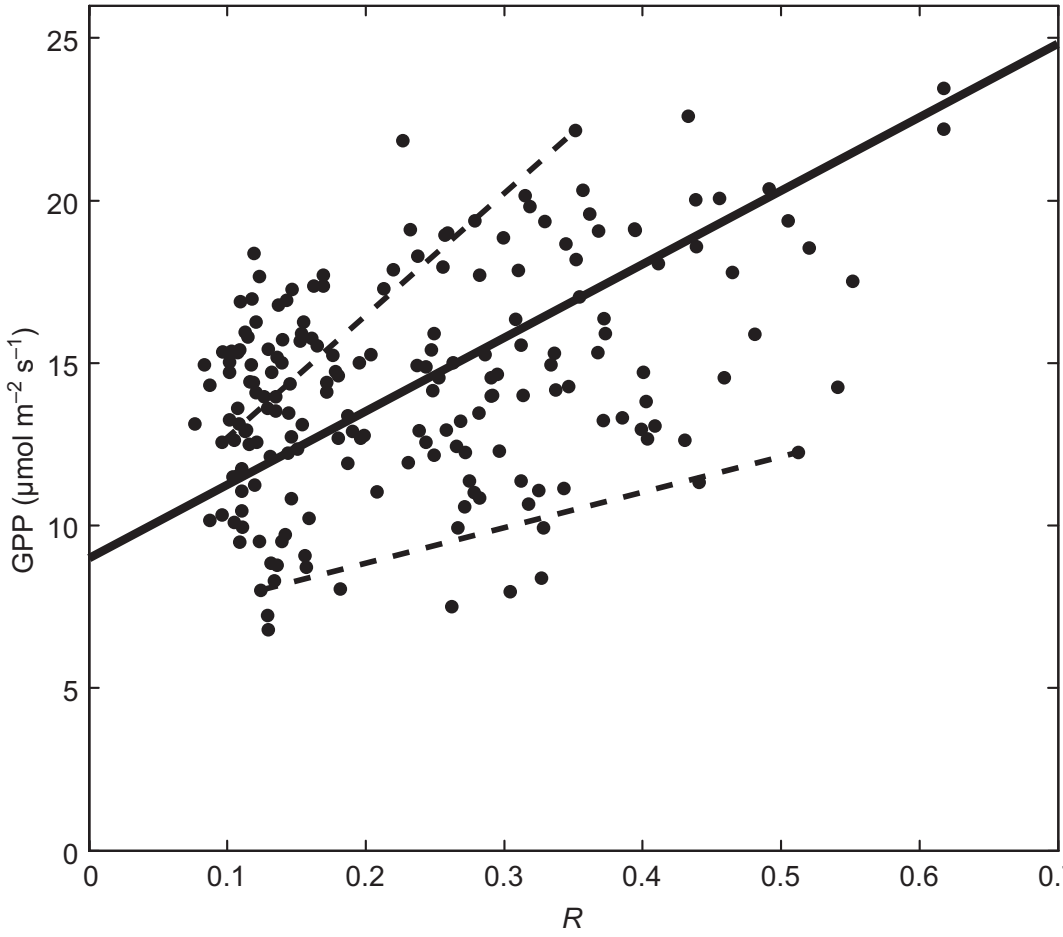
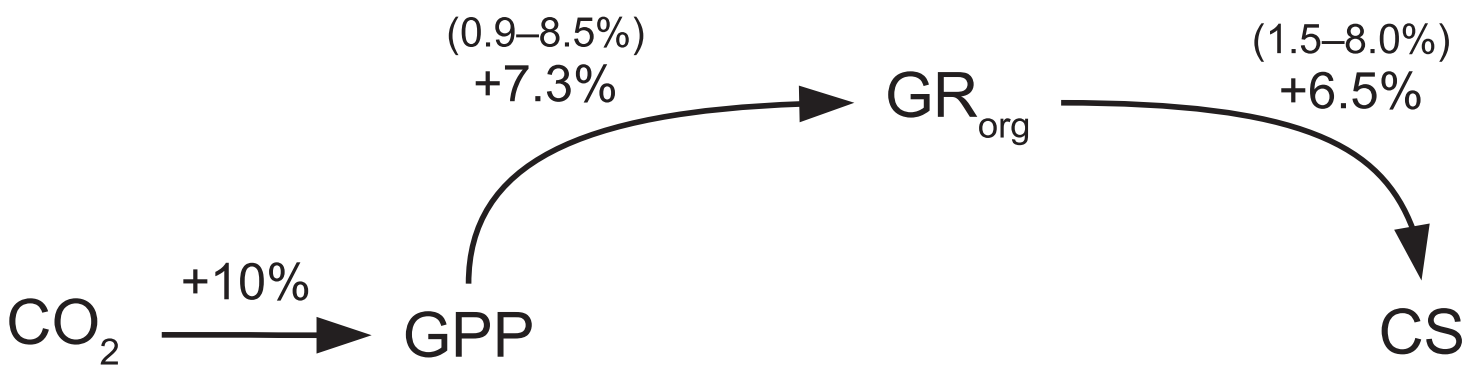


Fig. 6. GPP as a function of the ratio between diffuse and global radiation, *R*, averaged over the time period 08:00–20:00 in days when the corresponding average temperature is in the range 18–23 °C and average global radiation is in the range 500–600 W m^{-2}. The solid line shows the least-squares fit to the measurement points (slope = 22 μmol m^{-2} s^{-1}, r = 0.41, n = 393, p = 8.7 × 10^{-9}), and the dashed lines indicate the range of slopes used for upper- and lower-limit estimates. The data covers the years 2000–2010.

mated strengths of the individual steps varied considerably (Fig. 7). Our best estimate is that an increase of 10% in GPP, driven by the atmospheric CO_2 concentration increase, induces an additional increase of 3% in GPP due to the positive feedback. Our rough lower-limit and upper-limit estimates for this additional increase are 0.2% and 5%, respectively.

The strengths of feedbacks can be measured using feedback parameters or, alternatively, gains (e.g. Schwartz 2011). The gain G is defined as the change in the quantity of interest in the presence of the feedback divided by the corresponding change without the feedback. By using the values given in Fig. 7, the gain in GPP due to the feedback considered here is equal to



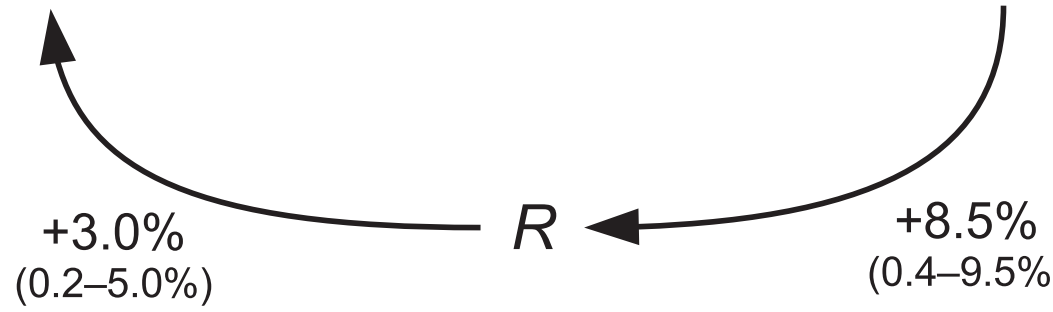


Fig. 7. Quantitative results of the GPP feedback loop obtained from observations made at SMEAR II station in Hyytiälä, Finland, during 1996–2011. For each step (Fig. 2), the percentage refers to the change in the second variable of that step that was related to the change in the first variable determined by the previous step. In practice, this procedure was done as follows: We first determined the median value of the first variable (the variable in the *x*-axis) and denoted this as X1. Then we took the percentage change of this variable from the previous step, increased the value of X1 accordingly, and obtained a new value denoted by X2. Next, we took the second variable (the variable in the *y*-axis) and calculated the values Y1 and Y2 that correspond to X1 and X2 based on the regression. The percentage increase in the second variable was then equal to 100% $\times$ (Y2 – Y1)/Y1. The initial change in GPP was assumed to be 10%. The values of the linear fit are taken from the regression analysis (solid lines in Figs. 3–6), while the upper and lower limits are based on rough visual estimates (dashed lines in Figs. 3–6).

1.3 (range = 1.02–1.5). While clearly weaker than the major physical feedbacks in the climate system, including the atmospheric water vapor and cloud feedback (Randall *et al*. 2007), the GPP feedback appears to be stronger than the few atmospheric chemistry-climate feedbacks estimated earlier (Raes *et al*. 2010).

Our approach has several features that might affect the estimated magnitude of the feedback loop. First, the relations involving GPP and R were determined in a specific temperature (18–23 °C) range and, in case of Step 4, in a narrow global radiation range. While necessary for separating the effects of GPP, temperature and radiation on BVOC emissions, it is clear that this procedure enhances the overall uncertainty of our analysis. Second, we were able to determine GR_{org} for nucleation event days only. There are strong indications that particle growth rates

on nucleation event days are usually somewhat higher than the particle growth rate averaged over all days at the same measurement site (Tunved *et al*. 2006, Väänänen *et al*. 2013). As a result, it is possible that the regression slope we obtained for Step 1 is too high. Third, the variables GPP, GR_{org}, CS and *R* are likely to respond to environmental changes over somewhat different time scales, so we may loose some essential information when using the temporal averages of 6 to 12 hours for these quantities. Fourth, the obtained strength of each step of the feedback is likely to depend on the time window over which the quantities related to this step were averaged. Fifth, we did not consider the potential influence of clouds and precipitation on the feedback loop.

Finally, it should be noted that most of the quantities involved in our analysis are affected not only by biogenic sources but also by anthropogenic activities (Arneth *et al*. 2010, Mahowald 2011, Makkonen *et al*. 2012a, Shindell *et al*. 2012, Spracklen and Rap 2013). Anthropogenic

effects are likely to change the strength of the observed relations, yet it is very difficult to estimate to which direction (increase or decrease) such changes would be. As an example, ozone represents an important threat to the forest growth in the northern hemisphere (Wittig *et al*. 2009). It has been estimated that current ozone levels reduce the forest carbon sequestration in some northern and central European countries by about 10% (Karlsson 2012), even though it is unclear how ozone affects growth of mature forests under field conditions. At our measurement site, ozone concentrations increased slightly during 1996–2011, influencing GPP and BVOC emissions opposite to the atmospheric CO_2 increase over the same time period. However, the moderate or even minor anthropogenic effect in Hyytiälä is implicitly taken into account in our analysis.

Conclusions and future outlook

We have made the first quantitative estimate regarding the terrestrial climate feedback loop that connects the increasing atmospheric carbon dioxide concentration, changes in gross primary production associated with carbon uptake by vegetation, organic aerosol formation in the atmosphere, and transfer of both diffuse and global radiation in cloud-free air. Our analysis was based on combining process-level

understanding with field measurements made in a boreal forest environment. The estimated strength of the feedback loop (gain 1.02–1.5) was found to be larger than the strengths of the few atmospheric chemistry–climate feedbacks estimated by other investigators using large-scale models (Raes *et al*. 2010), but smaller than the major climate feedbacks such as the water vapor feedback (Randall *et al*. 2007). We conclude that more detailed feedback studies in

a boreal forest environment are needed, and that similar studies should also be conducted in other terrestrial ecosystems.

Our analysis demonstrates the importance of making continuous, long-term, comprehensive field measurements when investigating the complicated couplings between the biosphere and atmosphere. However, in spite of having a very large data set, the observed relations connecting the various steps in the investigated feedback loop were statistically moderate. Improving the accuracy of our estimated feedback strength, and quantifying its uncertainty range, may therefore not be possible using field measurement data alone. We should think how to make complementary investigations on this feedback using e.g. satellite observations, and how to combine field and satellite data with corresponding data obtained from model simulations.

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