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A new feedback mechanism linking forests, aerosols, and climate

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Abstract. The possible connections between the carbon balance of ecosystems and aerosol-cloud-climate interactions play a significant role in climate change studies. Carbon dioxide is a greenhouse gas, whereas the net effect of atmospheric aerosols is to cool the climate. Here, we investigated the connection between forest-atmosphere carbon exchange and aerosol dynamics in the continental boundary layer by means of multiannual data sets of particle formation and growth rates, of CO_2 fluxes, and of monoterpene concentrations in a Scots pine forest in southern Finland. We suggest a new, interesting link and a potentially important feedback among forest ecosystem functioning, aerosols, and climate: Considering that globally increasing temperatures and CO_2 fertilization are likely to lead to increased photosynthesis and forest growth, an increase in forest biomass would increase emissions of non-methane biogenic volatile organic compounds and thereby enhance organic aerosol production. This feedback mechanism couples the climate effect of CO_2 with that of aerosols in a novel way.

1 Introduction

In 2001, the Intergovernmental Panel on Climate Change (IPCC) estimated the global and annual radiative forcing due to greenhouse gases and aerosols, along with natural changes associated with solar radiation. Emphasis was placed on the complexity of the combined direct and indirect forcing from both aerosols and gases as well as on the importance of improving our understanding of the role each of these three individual components plays in an integrated system. Such knowledge would reduce the uncertainty in current estimates of radiative forcing and enable a better prediction of the effects of anthropogenic activity on global change. The most important issue to resolve is how the different components affecting radiative forcing interact with one another. Here we propose a mechanism that couples the effect of CO_2 and aerosol particles on climate. This suggestion is based on connections among CO_2-induced climate change, increased vegetation productivity, emissions of non-methane biogenic volatile organic compounds (BVOCs), and their ability to form aerosol particles.

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2 Background

Aerosol particles affect the climate directly by reflecting or absorbing solar radiation and indirectly by acting as cloud condensation nuclei (CCN). Either way, aerosols reduce the amount of solar radiation reaching the Earth's surface. Among the key questions in reducing the uncertainties relating to radiative forcing of particles are how they are formed, how they grow from clusters of a few molecules to CCN sizes (>100 nm), and how they form cloud droplets. Formation of nanometre-sized aerosol particles and their subsequent growth to CCN sizes have been observed frequently in the continental boundary layer all around the world (Kulmala et al., 2004): from sub-arctic Lapland to the remote boreal forest in southern Finland (Mäkelä et al., 1997) and from central Europe (Birmili and Wiedensohler, 2000) to rural United Kingdom (Coe et al., 2000). Once formed, clouds influence Earth's radiation budget extensively by contributing to albedo and greenhouse effects. With global warming, cloud properties are likely to change due to warmer and moister conditions and also evidently due to increased aerosol particle concentrations from both primary (such as wind-generated sea spray) and secondary processes (from biogenically and anthropogenically influenced gas-to-particle conversion processes). The formation and growth of

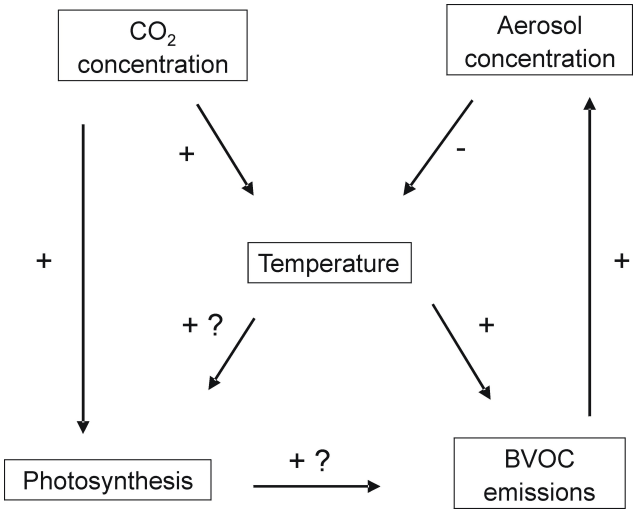

Fig. 1. Schematic figure of coupling of atmospheric CO_2 concentration, assimilation of carbon by vegetation productivity (ecosystem gross primary production GPP), emission of biogenic volatile organic compounds (BVOCs), and aerosol particle concentration with atmospheric temperature. Increased CO_2 concentration will increase temperature (+) and vegetation productivity (+). Increased temperature will enhance BVOC emissions (+) and probably also plant productivity (+?). Increased vegetation productivity may enhance BVOC emissions (+?). Increased BVOC emissions will enhance aerosol formation and growth and therefore also enhance aerosol and CCN concentrations (+). Enhanced aerosol and CCN concentrations will decrease temperature (−) due to increased reflection of sunlight from low clouds back to space. This results also in the increase of diffuse radiation, which has a positive influence on photosynthesis (Gu et al., 2003).

aerosol particles are related to the properties and transport of air masses as well as to biological activity and depend in a highly non-linear way on concentrations of nucleating and condensing vapours, temperature and relative humidity (Kulmala et al., 2000, 2001; Mäkelä et al., 2002; O'Dowd et al., 2002).

Terpenoids, among the most important BVOCs emitted by the vegetation, are known to lead to aerosol formation through rapid reactions with atmospheric oxidants such as ozone (O_3), hydroxyl (OH) radicals, and nitrate (NO_3) radicals (Seinfeld and Pandis, 1998). The products of these reactions possess low volatility due to various functional groups including, for instance, carboxylic acids. Therefore, they readily take part in gas-to-particle conversion processes (Atkinson, 2000; Calogirou et al., 1999). Plants synthesize terpenoids by two distinct pathways, the mevalonic acid (Bohlmann et al., 1998) and the 1-deoxy-D-xylulose-5-phosphate (Lichtenthaler, 1999). The first route is attributed to production of terpenoids (such as some monoterpenes) that are stored in specialized storage tissues and whose emissions follow the ambient temperature (Fuentes et al., 2000). The latter pathway, involved, for instance, in synthesis of isoprene and α-pinene, is coupled with a variety of environmental variables such as CO_2 concentration, temperature, or light intensity (Staudt and Bertin, 1998), and has been associated with carbon fixed immediately prior to synthesis (Loreto et al., 1996). Monoterpene emissions from boreal tree species such as Pinus sylvestris, Picea abies and Betula pubescens have also been attributed to light (Hakola et al., 2001; Janson, 1993; Schürmann et al., 1993). Recent measurements from Scots pine in Hyytiälä, southern Finland, showed a significant reduction in monoterpene emissions in dark conditions (Bäck et al., 2003) and suggest that both temperature and light play a role in controlling emission rates.

3 Proposed mechanism

Figure 1 shows the proposed mechanism and coupling among processes in forest ecosystems (vegetation productivity and BVOC emissions), aerosols, and climate. Photosynthesis drives ecosystem gross primary production (GPP), the difference between net ecosystem exchange of CO_2 (NEE) and total ecosystem respiration (TER). In the boreal zone, photosynthesis occurs predominantly in sunlight during the growing season (Hari and Mäkelä, 2003) and is inhibited in winter (Ottander et al., 1995). A negative feedback exists between atmospheric CO_2 concentrations and plant growth: Increasing CO_2 concentrations accelerate photosynthesis which in turn consumes more CO_2 (Lenton, 2000; Nemani et al., 2003). On the other hand, forest ecosystems also act as significant sources of atmospheric aerosols (Kulmala et al., 2001). Terrestrial vegetation contributes substantially to emissions of a variety of BVOCs (Fuentes et al., 2000) and newly formed particles in forested areas have been found to contain large amounts of organic material (O'Dowd et al., 2002). The ratio of BVOC emission to carbon assimilation is generally a few percent (Grace and Rayment, 2000; Guenther et al., 1995), and if increased CO_2 concentrations enhance vegetation productivity, we suggest that emission of aerosol-forming BVOCs may increase and possibly modify the aerosol particle formation routes (Kulmala et al., 2001).

In order to be able to test our hypothesis of the connection between aerosol formation and forest ecosystem activity, we analysed six years of field measurement data from Hyytiälä, southern Finland (61°51′ N, 24°17′ E, 181 m above sea level). Measurement of aerosol formation and growth (Kulmala et al., 2001), of surface fluxes as well as of meteorological variables and trace gases (temperature, radiation, O_3 etc.) have been performed continuously in Hyytiälä since 1996 (Vesala et al., 1998). Since the beginning of 2001, also BVOC concentrations in the air have been measured every third day (Hakola et al., 2003). We calculated formation rates for 3 nm particles (J3) as well as particle diameter growth rates (GR) directly from measured particle size distributions obtained with aerosol mobility spectrometers (Aalto et al., 2001). J3 varied between 0.02 and 2.3 $cm^{-3}\,s^{-1}$, with

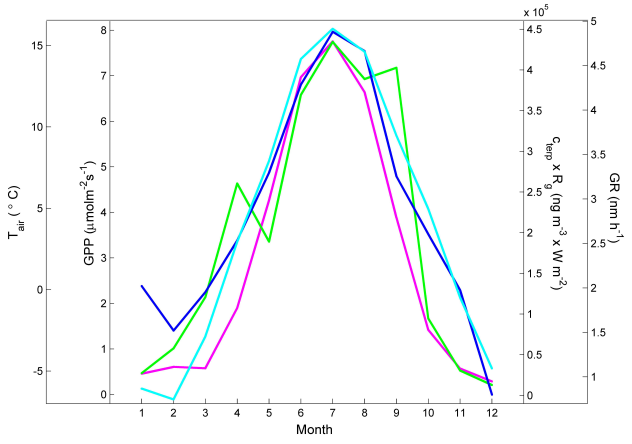

Fig. 2. Seasonal behaviour of GPP, growth rate, air temperature, and a proxy for BVOC oxidation products from OH-reactions. Growth rate (GR) for all events (dark blue), gross primary production (GPP) (pink), sum of mean monoterpene concentrations (c_{terp}) times global radiation (R_g) (green), and air temperature (T_{air}) (light blue). The points represent monthly means averaged over all years 1996–2001 (2001–2002 for terpenes).

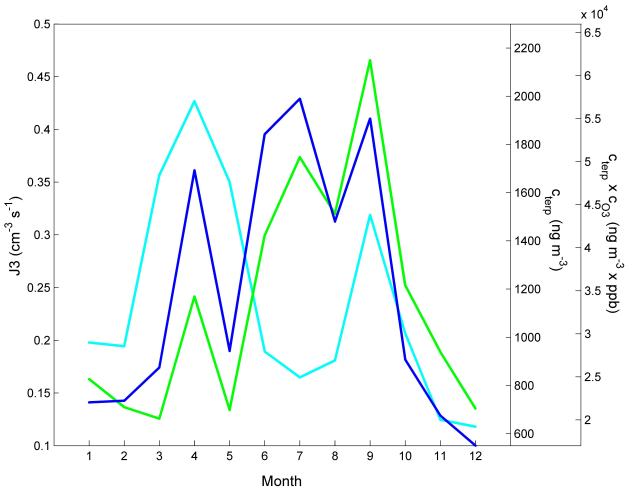

Fig. 3. Seasonal behaviour of formation rate, monoterpene concentrations, and a proxy for BVOC oxidation products from O_3-reactions. Formation rate of 3 nm-diameter particles J3 (light blue), mean monoterpene concentrations (c_{terp}) (green), sum of mean monoterpene concentrations times mean O_3 concentration. ($c_{\mathrm{terp}} \times c_{O_3}$) (dark blue). The points represent monthly means averaged over all years 1996–2001 (2001–2002 for terpenes).

a mean value of 0.36 $\mathrm{cm}^{-3}\,\mathrm{s}^{-1}$. The GR were obtained from an analysis of the size distribution evolution of 361 particle formation events observed from 1996 to 2001. The GR varied between 0.1 and 14.2 nm/h, with a mean of 3.1 nm/h. GPP was calculated as NEE-TER; NEE was measured by the eddy covariance technique (Markkanen et al., 2001; Suni et al., 2003), and TER was modelled on the basis of nighttime NEE measurements. For more details of the site and corresponding measurement techniques, see e.g. Kulmala et al. (2001) and Suni et al. (2003).

The growth rate of nucleation-mode particles has a clear maximum in summer. Figure 2 shows the monthly mean values, averaged over 6 years, for GPP, GR and air temperature (T_{air}). The figure also shows the monthly mean of the product of total monoterpene concentrations and global radiation (R_g), which is used here as a proxy for the formation of BVOC oxidation products from OH-reactions. Similar annual behaviour of the different parameters is clearly visible. In particular, GR peaks the same way (maximum in July) as does the proxy for the OH-oxidation products. The OH oxidation mechanism has recently been found to explain roughly 30–50% of the growth rates (Boy et al., 2003). Furthermore, GPP and T_{air} show almost exactly the same seasonal behaviour, suggesting that both light- and temperature-dependent BVOC emissions play an important role in the particle formation and growth process.

Figure 3 shows the observed J3 from the same data set, together with the product of mean monoterpene and ozone concentrations, which serves as a proxy for the products of the ozonolysis of BVOCs. The monthly patterns of these variables show some interesting similarities. In particular, both curves show similar peaks in spring and autumn. However, the summer peak visible in the ozonolysis product curve does not appear in J3. This is probably because particle formation typically occurs during cold air outbreaks within arctic or polar air masses (Kulmala et al., 2001; Nilsson et al., 2001), which are not common in summer months (June–August). In summer, sub-tropical air masses occur with high pre-existing aerosol concentrations, suppressing new-particle formation. The bimodal annual pattern of J3 is similar to the annual pattern of the frequency of nucleation events. For comparison, also the mean monoterpene concentration is shown in Fig. 3. This parameter exhibits its peak values significantly later during the year and the spring peak is less pronounced.

4 Discussion

The observed month-to-month patterns in Figs. 2 and 3 suggest firstly that the formation rate of new particles might also be linked with the rate of terpene reactions with O_3 (Fig. 3) and secondly that the overall GR is correlated with the rate of terpene reactions with OH (Fig. 2). The first of these observations could be explained by the formation of very low-volatility products from the ozonolysis reaction of certain terpenes as has been observed in several laboratory studies (Bonn and Moortgat, 2003; Hoffmann et al., 1998), and gains special relevance considering the increase of tropospheric ozone concentrations as a consequence of anthropogenic activities (Kanakidou et al., 2000). The second observation

demonstrates the general importance of BVOC oxidation on particle growth and, hence, on CCN formation.

At the global scale, BVOCs are emitted from vegetation with a rate of about 1.2×10^{15} gC per year (Guenther et al., 1995), which is ∼2% of the estimated global carbon assimilation by terrestrial ecosystems (Grace and Rayment, 2000). Under present conditions, increased CO_2 concentration will almost linearly increase CO_2 assimilation (Farquhar and von Caemmerer, 1982), which is likely to lead to increased BVOC emissions as well. The increased concentrations of BVOCs will then have an important effect on atmospheric chemistry, for example on O_3 formation (Seinfeld and Pandis, 1998), and particularly on the formation and growth of atmospheric aerosols (Kavouras et al., 1998). They will also enhance the condensational growth of small nuclei, and subsequently a larger fraction of aerosol particles will be able to grow to CCN sizes. Because of the uncertainties related to the coupling between ambient CO_2 concentrations and BVOC emissions, we can consider two extreme scenarios: Firstly, assuming that no coupling exists, doubling of atmospheric CO_2 concentration will not affect the global BVOC emission rates at all. Secondly, assuming complete coupling, doubling of atmospheric CO_2 concentration will also double the emission rates. As a moderate estimate, we assume below that the increase in BVOC emissions will be 10%. Note that we ignore the possible increase in BVOC emissions due to increased temperature, lengthened growing season, nitrogen fertilization, or increased leaf area index. These will make the increase stronger.

If we have 10% more condensable vapours due to an increase of 10% in BVOC emissions, the aerosol particles grow to CCN sizes in principle in roughly 10% shorter time and thus experience less scavenging by coagulation. However, since coagulation scavenging is a process leading to exponential decay in nucleation- mode number concentration and is strongly dependent on particle size (as a function of time), the effect of shorter growing times on the number concentration of CCN (c_{CCN}) is actually considerably larger – c_{CCN} may increase by much more than 10%. According to observed nucleation and growth events in a boreal forest (Kulmala et al., 2000, 2001), c_{CCN} will increase by a factor of 2 to 4 during an event (Kurtén et al., 2003). Since there are around 50 events per year (Kulmala et al., 2001), we can estimate that 30–60% of average total c_{CCN} stems from formation and growth events in the boreal forest region. Therefore, assuming a 10% increase in the CCN formation process, c_{CCN} will increase by 3–6%. This will subsequently increase the optical thickness of individual clouds by 1–2%, resulting in an increase in reflection of sunlight back to space. Note that the maximum increase in c_{CCN} may even be close to 100% if doubled atmospheric CO_2 concentration also leads to doubled BVOC emission rates (the extreme case of complete coupling). Then the optical thickness of individual clouds could increase even by 20%. According to a recent order-of-magnitude estimation, the contribution of boreal aerosol formation to the global radiative balance is -0.03 to $-1.1\,\mathrm{Wm}^{-2}$ (Kurtén et al., 2003). Therefore, assuming a 10% increase in c_{CCN}, the total contribution by CCN to the radiative balance will also increase by 10%. It is worth noting, though, that the cooling effect of a 10% increase in CCN is rather small compared to warming by a doubling of CO_2.

5 Conclusions

The results indicate two important connections in terms of seasonal variability, one between the growth rate of nucleation-mode aerosol particles and ecosystem gross primary production, and another between the formation rate of nucleation-mode particles and the ozonolysis of terpenoids. In addition, the seasonal pattern of particle growth rates is similar to that of the formation of oxidation products from terpene reactions with OH radicals. The proposed interaction between forest ecosystems, BVOC emissions, aerosol formation and clouds emphasizes the significance of forests on climate change. Thus forests, in addition to being sinks of CO_2, also act as sources for aerosol particles. For both of these reasons, increased forest growth leads to the slowing down of global warming. For more quantitative estimations, laboratory experiments, global climate modelling, and extensive international measurement campaigns are necessary.

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