



Phil. Trans. R. Soc. A doi:10.1098/rsta.2008.0201 Published online

# Boreal forests, aerosols and the impacts on clouds and climate

By Dominick V. Spracklen<sup>1,\*</sup>, Boris Bonn<sup>2</sup> and Kenneth S. Carslaw<sup>1</sup>

<sup>1</sup>Institute for Climate and Atmospheric Science, School of Earth and Environment, University of Leeds, Leeds LS2 9JT, UK <sup>2</sup>Institute for Atmospheric and Environmental Sciences, Frankfurt University, 60438 Frankfurt/Main, Germany

Previous studies have concluded that boreal forests warm the climate because the cooling from storage of carbon in vegetation and soils is cancelled out by the warming due to the absorption of the Sun's heat by the dark forest canopy. However, these studies ignored the impacts of forests on atmospheric aerosol. We use a global atmospheric model to show that, through emission of organic vapours and the resulting condensational growth of newly formed particles, boreal forests double regional cloud condensation nuclei concentrations (from approx. 100 to approx. 200 cm<sup>-3</sup>). Using a simple radiative model, we estimate that the resulting change in cloud albedo causes a radiative forcing of between -1.8 and -6.7 W m<sup>-2</sup> of forest. This forcing may be sufficiently large to result in boreal forests having an overall cooling impact on climate. We propose that the combination of climate forcings related to boreal forests may result in an important global homeostasis. In cold climatic conditions, the snow-vegetation albedo effect dominates and boreal forests warm the climate, whereas in warmer climates they may emit sufficiently large amounts of organic vapour modifying cloud albedo and acting to cool climate.

Keywords: boreal forests; climate feedbacks; aerosols; cloud condensation nuclei; climate mitigation; new particle formation

## 1. Introduction

There is growing interest in the use of forest management to mitigate anthropogenic climate change. Forests store carbon within vegetation and soil, so increasing forest area (through reforestation or reduced deforestation) and increasing carbon storage per unit area of forest (e.g. through sustainable logging practices) can help to slow the increase of atmospheric carbon dioxide concentrations. However, climate mitigation is complicated by the multiple ways in which forests impact climate. In addition to impacts on the global carbon cycle, forests can alter the composition of the atmosphere through the emission of gas-phase and aerosol species (biogeochemical effects) and can modify land-surface properties (biophysical effects). In this paper, we use a global

1

\*Author for correspondence (d.spracklen@see.leeds.ac.uk).

One contribution of 10 to a Triennial Issue 'Earth science'.

atmospheric model to quantify the impact of biogenic boreal forest emissions on atmospheric aerosol, cloud properties and climate through the first aerosol indirect effect.

## 2. Boreal forests and climate

Forests have complex interactions with the atmosphere. They modify the surface-atmosphere exchange of energy, momentum, water, carbon dioxide and other trace gas and aerosol species (figure 1). Through these interactions, they impact regional and global climates. However, the impacts on large-scale climate are difficult to observe directly and predictions generally rely on global numerical models. Early studies, made using general circulation models, accounted only for the biophysical effects of the forest (e.g. Dickinson & Henderson-Sellers 1988), whereas later studies evaluated both biophysical and carbon cycle impacts. While these studies predict that tropical forests cause climate cooling, due to large carbon storage combined with large evapotranspiration (ET) promoting low-level cloud cover (e.g. Bala et al. 2007: Sampaio et al. 2007), the impact of boreal forests on climate is less certain. Boreal forests (location shown in figure 2) have a dark canopy (with low albedo) that obscures the snow-covered ground (with high albedo), absorbs sunlight and warms the climate (known as the snow-vegetation albedo effect) (Thomas & Rowntree 1992; Chalita & Le Treut 1994). This effect is predicted to dominate over the cooling from ET and carbon storage, meaning that boreal forests warm the climate (Bonan et al. 1992; Betts 2000; Claussen et al. 2001; Bala et al. 2007). Bala et al. (2007) predict that boreal forests warm global climate by  $0.8^{\circ}$ C. Randerson *et al.* (2006) used observations of the radiation balance over a chronosequence of forest fire burn scars combined with climate model analysis to suggest that the radiative forcing of mature boreal forest compared with forest over an 80-year fire cycle was a warming of  $2.3 \pm 2.2 \text{ W m}^{-2}$ .

However, these previous studies have not been able to include the impact of boreal forests on atmospheric particles and clouds. Boreal forest regions contain very few anthropogenic sources of pollution, so emissions from the forest may be an important source of particles (Andreae 2007) and may significantly alter the regional radiation balance.

#### 3. Boreal forests and aerosols

Boreal forests modify atmospheric particles in several ways. Vegetation emits biogenic volatile organic compounds (BVOCs) that can be oxidized in the atmosphere to form products with low enough vapour pressure to condense on existing aerosol particles, forming secondary organic aerosol (SOA). This SOA is an important component of the particulate load in many environments (Zhang *et al.* 2007) including the boreal forest. The most important BVOCs emitted by boreal forests are monoterpenes ( $C_{10}H_{16}$ ), with the strength of emission depending on the tree species and varying according to temperature and light among other variables.

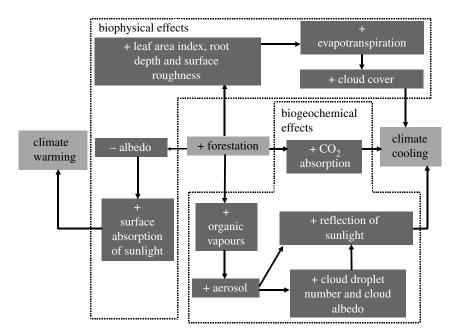


Figure 1. A simplified schematic of the impacts of increased forestation on climate.



Figure 2. Boreal forests (dark grey) are an extensive ecosystem covering over 15 million  $\mathrm{km}^2$  of northern Siberia, North America and northern Europe and constitute about one-third of global forest cover.

Vegetation can also emit particles directly into the atmosphere. Primary biological aerosol particles include spores, fungi and leaf matter. Very little is known about the importance of these particles for atmospheric composition and climate. They may dominate the large aerosol size mode over forested regions and may be important ice nuclei impacting rain formation (Diehl *et al.* 2001). However, owing to very limited knowledge about sources, for the rest of this work, we do not consider primary biological aerosol.

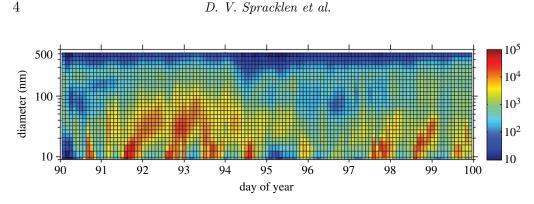


Figure 3. Observed aerosol size distribution  $(dN/d \log Dp \text{ in cm}^{-3})$  at the Hyytiälä observatory  $(61^{\circ}51' \text{ N}, 24^{\circ}7' \text{ E})$  located in extensive boreal forest in southern Finland during spring 2003.

A ubiquitous feature at boreal forest sites is the regular occurrence of new particle formation events (e.g. Kulmala *et al.* 2001; Tunved *et al.* 2003). New particle formation is the conversion of gas-phase species into particles and is observed at many continental sites around the world (Kulmala *et al.* 2004*a*). Figure 3 shows new particle formation events occurring most days at a Finnish boreal forest site. The newly formed particles grow rapidly to above 70 nm in diameter where they act as effective cloud condensation nuclei (CCN). Analysis of the aerosol size distribution before and after such formation events shows that they contribute substantially to local CCN concentrations (Lihavainen *et al.* 2003; Kerminen *et al.* 2005; Laaksonen *et al.* 2005). Spracklen *et al.* (2008) used an atmospheric model combined with an empirical particle formation rate to show that such particle formation can increase global springtime CCN concentrations (at 0.2% supersaturation) by 3–20 per cent.

The role that boreal forests play in instigating particle formation is unclear primarily because the formation mechanism is not well understood. Analysis of observations, including those over boreal forests, suggests that sulphuric acid plays a key role, with the formation rate of 1 nm molecular clusters  $(J_1)$  being proportional to the gas-phase sulphuric acid concentration to the power M, where M is between 1 and 2,

$$J_1 = k [H_2 SO_4]^M,$$
 (3.1)

and k is a rate coefficient that has been observed to vary spatially and temporally by several orders of magnitude (Kulmala *et al.* 2006; Sihto *et al.* 2006; Riipinen *et al.* 2007; Kuang *et al.* 2008). The cause of the variability in k and M is not well understood. Nevertheless, at an empirical level, this mechanism (with  $k=2\times10^{-6}$  s<sup>-1</sup> and M=1) can explain observed particle formation events over boreal regions (Sihto *et al.* 2006; Spracklen *et al.* 2006).

Although the role of forest emissions in instigating formation events is unclear, it is well understood that the oxidized BVOCs can increase particle growth rates and thereby influence the production of CCN from the newly formed particles. In remote continental environments, sulphuric acid explains only a fraction (generally less than 10%) of the observed growth of newly formed particles (Kulmala *et al.* 2001, 2004*a*; Boy *et al.* 2003, 2005; Tunved *et al.* 2006*b*), with the rest being attributed to SOA material from organic vapours emitted from the forest (O'Dowd *et al.* 2002; Allan *et al.* 2006; Tunved *et al.* 2006*a*; Laaksonen *et al.* 2008).

#### 4. Estimating the contribution of boreal forests to atmospheric aerosol

Tunved *et al.* (2006*a*) studied the evolution of aerosol in air that was transported from clean oceanic regions across the boreal forest to field stations in Finland. They showed that the aerosol mass increased with the time that the air had spent over forested regions, suggesting the importance of forest emissions. Tunved *et al.* (2008) calculate that boreal forests contribute 12–50 per cent of observed aerosol mass over Scandinavia and can sustain CCN concentrations of approximately 200 cm<sup>-3</sup>, double that observed in marine air masses. Kurten *et al.* (2003) used observations of new particle formation events made at the Hyytiälä field station in southern Finland to estimate that aerosol formation leads to a cooling forcing of between -5 and -14 W m<sup>-2</sup> of forest.

We use the GLOMAP global aerosol model (Spracklen *et al.* 2005a,b) to further study the influence of boreal terpene emissions on aerosol properties. GLOMAP is an extension to the TOMCAT three-dimensional chemical transport model (Chipperfield 2006). The model is forced with European Centre for Medium-Range Weather Forecasts analyses and uses a horizontal resolution of  $2.8^{\circ} \times 2.8^{\circ}$  and 31 vertical levels between the surface and 10 hPa. The model simulates size and composition-resolved aerosol using two externally mixed distributions, each described by a two-moment sectional scheme with 20 sections spanning 3 nm to  $25 \,\mu m$  dry diameter. One distribution, representing freshly emitted primary carbonaceous aerosol, contains organic carbon and elemental carbon, is treated as hydrophobic and is not wet scavenged. The other distribution contains sulphate, sea salt, elemental carbon and organic carbon, is hydrophilic and is wet scavenged. The model includes both natural (e.g. oceanic dimethyl sulphide, volcanic  $SO_2$ , wildfire carbonaceous aerosol, sea salt, vegetation emissions of monoterpenes) and anthropogenic (e.g. fossil fuel emissions of  $SO_2$  and carbonaceous and sulphate aerosol) emissions of aerosols and aerosol precursor gases (Spracklen et al. 2006). We assume that the firststage oxidation products of monoterpenes form hydrophilic SOA with a yield of 13 per cent, similar to that observed over boreal regions (Tunved *et al.* 2006*a*).

We include new particle formation events in the boundary layer according to the observed nucleation rate (equation (3.1)) with  $k=2\times10^{-6}$  s<sup>-1</sup> and M=1, which matches observed aerosol concentrations at many continental sites (Spracklen *et al.* 2008) including a site within the Finnish boreal forest (Spracklen *et al.* 2006). Above the boundary layer, we use the binary homogeneous nucleation from Kulmala *et al.* (1998).

We ran two model simulations: a control simulation and a simulation where emissions of monoterpenes from boreal forest vegetation are modified to that of boreal tundra (C3 Arctic grass). The two model runs are identical in all other respects. For example, we do not modify the wet and dry deposition of aerosol and gases. Standard monthly mean emissions of monoterpenes are from Guenther *et al.* (1995). Monoterpene emissions from C3 Arctic grasslands are estimated to be 2 per cent of those from boreal forest vegetation (table 1).<sup>1</sup>

<sup>1</sup>Vegetation emissions of BVOCs at a specific temperature and light intensity are calculated using F = ED (Guenther *et al.* 1995), where *F* is the emission flux; *E* is the plant type emission capacity; and *D* is the foliar density, calculated by dividing the (two-sided) leaf area index (LAI) by the specific leaf area. Values chosen for the different ecosystems are shown in table 1.

Table 1. Monoterpene emission capacity, specific leaf area and leaf area index for boreal evergreen and C3 Arctic grass.

plant functional type	monoterpene emission capacity, $E (\mu g C g^{-1} dry)$ foliar mass $h^{-1}$ ; Levis <i>et al.</i> 2003)	dry foliar mass; Kucharik <i>et al</i> .	leaf area index $(m^2 m^{-2}; Chen et al. 2005)$	D (g dry foliar	emission flux, $F$ ( $\mu$ g C m <sup>-2</sup> h <sup>-1</sup> )
boreal ever-	2	0.00625	7	1120	
green C3 grass,	0.1	0.01	2	200	2240
Arctic		0.01	-		20

Changing from forest to C3 grassland emissions in boreal regions results in only approximately 20 per cent reduction in global summertime monoterpene emissions since global emissions are dominated by tropical vegetation. However, as we show, the local effects can be substantial.

Figure 4 shows the boundary-layer (BL) CCN concentrations (at 0.2% supersaturation, corresponding to an activation diameter of approx. 70 nm) in the model runs with forest terpene emissions and with forest terpene emissions replaced with those from Arctic grassland. In the grassland simulation, summertime (June–August) mean CCN concentrations north of 60° N are relatively low at approximately 120 cm<sup>-3</sup>. This is much lower than concentrations simulated at mid-latitudes (mean of approx. 800 cm<sup>-3</sup> between 30 and  $60^{\circ}$  N) and reflects the lack of pollution sources in boreal regions. Boreal forest terpene emissions increase CCN concentrations north of  $60^{\circ}$  N by a factor of almost 2, to 230 cm<sup>-3</sup>, which is similar to the estimate of Tunved *et al.* (2008). This increase in CCN is due to oxidation products of terpene emissions condensing on and contributing to the growth of small particles, which then grow to large enough sizes to act as CCN.

This large impact of boreal forest terpene emissions on CCN concentrations is amplified by particle formation. Particle formation events provide many new particles in the BL, which can grow through condensation of SOA to become CCN. In a sensitivity scenario where we switch off particle formation events in the BL, boreal terpene emissions increase CCN concentrations by only approximately 50 per cent, from 110 to 160 cm<sup>-3</sup>. Therefore, particle formation events approximately double the impact of forest terpenes on CCN concentrations.

Here we have assumed, as observed, that sulphuric acid controls the particle formation rate, which is therefore largely controlled by anthropogenic emissions of sulphur dioxide. Laboratory studies have shown that nucleation rates are enhanced by the presence of organic species (Zhang *et al.* 2004; Verheggen *et al.* 2007), and the formation of organo-sulphate clusters has been suggested as an ambient particle formation mechanism (Bonn *et al.* 2008; Boy *et al.* 2008). Bonn *et al.* (2008) showed that a mechanism involving the oxidation products of biogenic terpenes could explain particle formation events observed at a boreal forest site in Finland. If biogenic emissions do play an important role in the particle formation rate, CCN concentrations may be more tightly coupled to biogenic emissions than was calculated here.

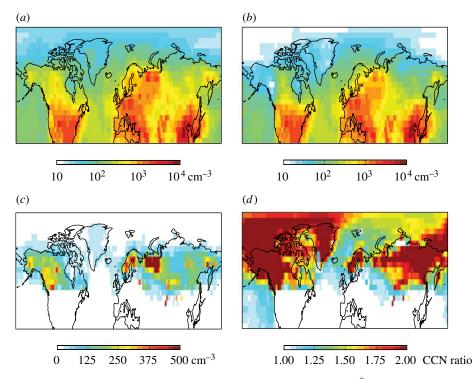


Figure 4. Summertime (June–August) mean concentrations (cm<sup>-3</sup>) of CCN (0.2% supersaturation) in simulations with (a) boreal forest terpene emissions, (b) boreal forest terpene emissions replaced with emissions from C3 Arctic grasslands (approx. 2% of forest emissions), (c) CCN with boreal emissions minus CCN with Arctic grassland emissions, and (d) ratio of CCN with boreal forest terpene emissions to CCN with Arctic grassland emissions.

Box 1. Aerosol radiative forcing.

Aerosols can modify the Earth's radiative balance through the aerosol direct and indirect effects.

—Aerosol direct effect. Aerosols directly scatter and absorb radiation. The scattering of radiation causes atmospheric cooling, whereas absorption can cause atmospheric warming.
 —Aerosol indirect effect. Aerosols modify the properties of clouds through a subset of the aerosol population called cloud condensation nuclei (CCN). Increased CCN concentrations lead to increased cloud droplet number concentrations (CDNC). A greater number of cloud droplets leads to increased cloud albedo, increased light scattering and radiative cooling (first indirect effect). Increased CDNC also leads to reduced precipitation efficiency and increased lifetime of the cloud (second indirect effect).

## 5. Climate forcing from forest-derived particles

The change in aerosol properties due to boreal forest terpenes will lead to an impact on climate through both direct and indirect aerosol radiative forcing (see box 1). The direct forcing from SOA will probably lead to a cooling (Chung & Seinfeld 2002); however, this effect is poorly quantified due partly to uncertainty in the optical properties of SOA. The increased CCN concentrations that

we quantified in the last section will increase cloud droplet number concentrations (CDNC) and increase the albedo (first indirect effect) and the lifetime of clouds (second indirect effect). The second indirect effect is poorly understood, so here we focus on estimating the first aerosol indirect effect from boreal forest emissions.

We calculate the change in cloud albedo  $(\Delta R_c)$  due to the change in cloud droplet number concentrations ( $\Delta CDNC$ ) according to Seinfeld & Pandis (1998) as

$$\Delta R_{\rm c} = (1/3)R_{\rm c}(1-R_{\rm c})\Delta \text{CDNC/CDNC}.$$
(5.1)

We assume that  $\Delta$ CDNC is equal to the change in CCN, an assumption that is applicable at low CCN concentrations (less than approx. 800 cm<sup>-3</sup>). Using (5.1), we calculate that boreal forest terpenes lead to a 3–8 per cent increase in cloud albedo, where the uncertainty in our estimate is due to the observed range of cloud albedo in boreal regions, which varies between 0.1 and 0.8 (Betts *et al.* 2007).

We use a single reflection radiation model to estimate the short-wave forcing from this change in cloud albedo. This simple model assumes a layer of stratocumulus cloud over an ocean surface with zero albedo (that is, completely absorbing). Since cloud forcing decreases with increasing surface albedo, this assumption will overestimate our calculated forcing where the cloud is over land or ice. The change in short-wave forcing,  $\Delta F_c$ , is calculated according to Seinfeld & Pandis (1998) as

$$\Delta F_{\rm c} = -F_0 A_{\rm c} T_{\rm a}^2 \Delta R_{\rm c}, \qquad (5.2)$$

where  $T_{\rm a}$  is the fractional atmospheric transmission of short-wave radiation above the cloud layer (assumed to be 0.76) and  $F_0$  is the top-of-atmosphere incoming solar radiation (assumed to be 204 W m<sup>-2</sup>, which is the annual mean incoming radiation for 60–90° N). An average fractional cloud cover ( $A_c$ ) of 0.65 (range approx. 0.55–0.75) for the boreal forest zone is calculated from 1987 to 2004 summertime (June–August) cloud fraction from the International Satellite Cloud Climatology Project (http://isccp.giss.nasa.gov/index.html). From (5.2), we calculate a regionally averaged (60–90° N) forcing of between -1.8 and -6.7 W m<sup>-2</sup> of forest, where the uncertainty in our forcing estimates is due to the range of values we assume for cloud albedo and fractional cloud cover. Our calculated forcing is smaller than that calculated by Kurten *et al.* (2003), possibly because they attributed all observed particle formation events to forests. We have calculated the impact of removing the forest source of terpenes, but have allowed for the fact that the nucleation events themselves are caused by emissions from non-forest sources.

Our estimated cooling from the aerosol indirect effect is sufficiently large to potentially change our understanding of the net climate forcing from boreal forests. Randerson *et al.* (2006) calculated the combined forcing from carbon cycle and biophysical radiative effects to be a warming of  $+2.3 \text{ W m}^{-2}$ . Although these forcing estimates are not directly comparable, combining our estimated cooling from the aerosol indirect effect with the estimate of Randerson *et al.* (2006) potentially gives boreal forests a net cooling impact on climate.

## 6. Boreal forest climate feedbacks

Increased temperature drives increased BVOC emissions (e.g. Guenther *et al.* 1995), which can drive faster particle growth rates, greater CCN concentrations and increased aerosol radiative cooling. A feedback between boreal forests, BVOC emissions, aerosols, clouds and climate is therefore possible (Kulmala *et al.* 2004*b*). Tunved *et al.* (2008) suggest that a 1.4°C increase in temperature would increase CCN concentrations over Scandinavia by 8 per cent and a 5.8°C increase in temperature would increase CCN concentrations by 40 per cent.

While the feedback between forests, monoterpene emissions, aerosols and climate has been reported previously, the combination of different boreal forest forcings could lead to some interesting and so far unexplored behaviour.

The magnitudes of the different boreal forest forcings vary with temperature, resulting in an overall forcing that also depends on temperature.<sup>2</sup> During cold climatic periods:

- snow will lie on the ground for most of the spring and summer so the warming from the snow-vegetation albedo effect will be large and
- monoterpene emissions will be small and so the cooling from the boreal forestaerosol-cloud albedo effect will be small.

Therefore, in a cold climate, the snow-vegetation albedo effect is likely to dominate and the overall climate forcing from boreal forests is likely to be a warming. The retraction of boreal forests has been shown to provide a positive feedback for glaciation (Meissner *et al.* 2003). As the climate warms:

- snow will melt earlier in the season and so the warming from the snow-vegetation albedo effect decreases and
- monoterpene emissions will increase and so the cooling from the boreal forestaerosol-cloud albedo effect also increases.

With increasing temperature, the net warming from boreal forests will decrease (figure 5). At some temperature, the forest–aerosol–cloud albedo effect may start to dominate and the overall climate forcing from boreal forests may be a cooling. Through these linked mechanisms, boreal forests may help to stabilize regional and global temperatures. Under cold climatic conditions, boreal forests may act to warm climate; whereas under warm climate conditions, they may act to cool climate.

### 7. Summary

We have made the first global model study of the impacts of terpene emissions from boreal forests on CCN concentrations. Boreal forest regions are relatively far from major anthropogenic sources of pollution and so forest-derived particles can be important in controlling the regional aerosol budget. We use simulations where we reduce the terpene emissions from boreal forest vegetation equivalent

 $^{2}$  For simplicity, we have here assumed that forest carbon storage and evapotranspiration do not vary with temperature, although this is not likely to be the case and will further complicate the combined feedbacks.



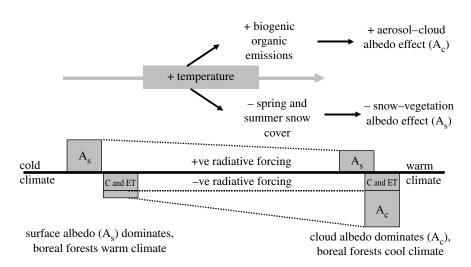


Figure 5. Combined boreal forest climate feedbacks. In this simplified schematic, we assume that carbon storage (C) and evapotranspiration (ET) effects do not change with changing climate.

to that of Arctic tundra to show that forest emissions approximately double the regional (north of  $60^{\circ}$  N) CCN concentrations, from approximately 100 to approximately 200 cm<sup>-3</sup>.

We estimate that this change in CCN concentrations results in an indirect aerosol radiative forcing of between -1.8 and -6.7 W m<sup>-2</sup> of forest. This is of comparable magnitude, but opposite sign, to previous estimates of the surface albedo effect of boreal forests (e.g. Kurten *et al.* 2003; Randerson *et al.* 2006) and may be large enough to modify our understanding of the net impact of boreal forests on climate.

This work has important policy implications. Previous studies have suggested that climate mitigation through forest expansion in the boreal zone would warm climate. Our study questions this conclusion. Climate model studies that comprehensively evaluate all of the influences of forests on climate are now required. Our study focused on the impacts of forest terpenes on aerosol, but other biogenic aerosol and trace gas sources may also be important.

We propose that a combination of climate forcings may result in boreal forests acting to help stabilize regional and global temperatures. During cold climatic periods, a dominant snow-vegetation albedo forcing results in boreal forests warming climate. In warm climatic periods, this forcing becomes less important and the forest-aerosol-cloud albedo forcing may dominate, resulting in boreal forests cooling climate. This impact of boreal forest on aerosol and clouds represents an important climate feedback process that must be included in climate models in order to make realistic predictions.

D.V.S. is funded through APPRAISE. We acknowledge a short visit grant from the European Science Foundation 'Volatile Organic Compounds in the Biosphere–Atmosphere System' (VOCBAS). We acknowledge the two anonymous referees.

#### References

Allan, J. D. et al. 2006 Size and composition measurements of background aerosol and new particle growth in a Finnish forest during QUEST 2 using an aerodyne aerosol mass spectrometer. Atmos. Chem. Phys. 6, 315–327. See http://www.atmos-chem-phys.net/6/315/2006/. Andreae, M. O. 2007 Aerosols before pollution. *Science* **315**, 50–51. (doi:10.1126/science.1136529)

- Bala, G., Caldeira, K., Wickett, M., Phillips, T. J., Lobell, D. B., Delire, C. & Mirin, A. 2007 Combined climate and carbon-cycle effects of large-scale deforestation. *Proc. Natl Acad. Sci.* USA 104, 16. (doi:10.1073/pnas.0608998104)
- Betts, A. K., Desjardins, R. L. & Worth, D. 2007 Impact of agriculture, forest and cloud feedbacks on the surface energy budget in BOREAS. Agric. Forest Meteorol. 142, 156–169. (doi:10.1016/ j.agrformet.2006.08.020)
- Betts, R. A. 2000 Offset of the potential carbon sink from boreal forestation by decreases in surface albedo. *Nature* **408**, 187–190. (doi:10.1038/35041545)
- Bonan, G. B., Pollard, D. & Thompson, S. L. 1992 Effects of boreal forest vegetation on global climate. *Nature* 359, 716–718. (doi:10.1038/359716a0)
- Bonn, B., Kulmala, M., Riipinen, I., Sihto, S.-L. & Ruuskanen, T. M. 2008 How biogenic terpenes govern the correlation between sulfuric acid concentrations and new particle formation. J. Geophys. Res. 113, D12209. (doi:10.1029/2007JD0909327)
- Boy, M., Rannik, Ü., Lehtinen, K. E. J., Tarvainen, V., Hakola, H. & Kulmala, M. 2003 Nucleation events in the continental boundary layer: long-term statistical analyses of aerosol relevant characteristics. J. Geophys. Res. 108, 4667. (doi:10.1029/2003JD003838)
- Boy, M. et al. 2005 Sulphuric acid closure and contribution to nucleation mode particle growth. Atmos. Chem. Phys. 5, 863–878. See http://www.atmos-chem-phys.net/5/863/2005/.
- Boy, M. et al. 2008 New particle formation in the Front Range of the Colorado Rocky Mountains. Atmos. Chem. Phys. 8, 1577–1590. See http://www.atmos-chem-phys.net/8/1577/2008/.
- Chalita, S. & Le Treut, H. 1994 The albedo of temperate and boreal forest and the Northern Hemisphere climate: a sensitivity experiment using the LMD GCM. *Clim. Dyn.* 10, 231–240. (doi:10.1007/BF00208990)
- Chen, X., Vierling, L., Deering, D. & Conley, A. 2005 Monitoring boreal forest leaf area index across a Siberian burn chronosequence: a MODIS validation study. Int. J. Remote Sens. 26, 5433–5451. (doi:10.1080/01431160500285142)
- Chipperfield, M. P. 2006 New version of the TOMCAT/SLIMCAT off-line chemical transport model: intercomparison of stratospheric tracer experiments. Q. J. R. Meteorol. Soc. 132, 1179–1203. (doi:10.1256/qj.05.51)
- Chung, S. H. & Seinfeld, J. H. 2002 Global distribution and climate forcing of carbonaceous aerosols. J. Geophys. Res. Atmos. 107, 4407. (doi:10.1029/2001JD001397)
- Claussen, M., Brovkin, V. & Ganopolski, A. 2001 Biogeophysical versus biogeochemical feedbacks of large-scale land cover change. *Geophys. Res. Lett.* 28, 1011–1014. (doi:10.1029/2000 GL012471)
- Dickinson, R. E. & Henderson-Sellers, A. 1988 Modeling tropical deforestation—a study of GCM land surface parametrizations. Q. J. R. Meteorol. Soc. 114, 439. (doi:10.1002/qj.49711448009)
- Diehl, K., Quick, C., Matthias-Maser, S., Mitra, S. K. & Jaenicke, R. 2001 The ice nucleating ability of pollen. Part I: Laboratory studies in deposition and condensation freezing modes. *Atmos. Res.* 58, 75–87. (doi:10.1016/S0169-8095(01)00091-6)
- Guenther, A. et al. 1995 A global model of natural volatile organic compound emissions. J. Geophys. Res. Atmos. 100, 8873–8892. (doi:10.1029/94JD02950)
- Kerminen, V.-M., Lihavainen, H., Komppula, M., Viisanen, Y. & Kulmala, M. 2005 Direct observational evidence linking atmospheric aerosol formation and cloud droplet activation. *Geophys. Res. Lett.* 32, L14803. (doi:10.1029/2005GL023130)
- Kuang, C., McMurry, P. H., McCormick, A. V. & Eisele, F. L. 2008 Dependence of nucleation rates on sulfuric acid vapor concentration in diverse atmospheric locations. J. Geophys. Res. 113, D10209. (doi:10.1029/2007JD009253)
- Kucharik, C. J. et al. 2000 Testing the performance of a dynamic global ecosystem model: water balance, carbon balance, and vegetation structure. Global Biogeochem. Cycles 14, 795–825. (doi:10.1029/1999GB001138)
- Kulmala, M., Laaksonen, A. & Pirjola, L. 1998 Parameterizations for sulfuric acid/water nucleation rates. J. Geophys. Res. Atmos. 103, 8301–8307. (doi:10.1029/97JD03718)

- Kulmala, M. et al. 2001 Overview of the international project on biogenic aerosol formation in the boreal forest (BIOFOR). Tellus B 53, 324–343. (doi:10.1034/j.1600-0889.2001.d01-24.x)
- Kulmala, M., Vehkamäki, H., Petäjä, T., Dal Maso, M., Lauri, A., Kerminen, V. M., Birmili, W. & McMurry, P. H. 2004a Formation and growth rates of ultrafine atmospheric particles: a review of observations. J. Aerosol. Sci. 35, 143–176. (doi:10.1016/j.jaerosci.2003.10.003)
- Kulmala, M. et al. 2004b A new feedback mechanism linking forests, aerosols, and climate. Atmos. Chem. Phys. 4, 557–562. See http://www.atmos-chem-phys.net/4/557/2004/.
- Kulmala, M., Lehtinen, K. E. J. & Laaksonen, A. 2006 Cluster activation theory as an explanation of the linear dependence between formation rate of 3 nm particles and sulphuric acid concentration. Atmos. Chem. Phys. 6, 787–793. See http://www.atmos-chem-phys.net/6/ 787/2006/.
- Kurten, T. et al. 2003 Estimation of different forest-related contributions to the radiative balance using observations in southern Finland. Boreal Environ. Res. 8, 275–285.
- Laaksonen, A., Hamed, A., Joutsensaari, J., Hiltunen, L., Cavalli, F., Junkermann, W., Asmi, A., Fuzzi, S. & Facchini, M. C. 2005 Cloud condensation nucleus production from nucleation events at a highly polluted region. *Geophys. Res. Lett.* 32, L06812. (doi:10.1029/ 2004GL022092)
- Laaksonen, A. et al. 2008 The role of VOC oxidation products in continental new particle formation. Atmos. Chem. Phys. 8, 2657–2665. See http://www.atmos-chem-phys.net/8/2657/ 2008/.
- Levis, S., Wiedinmyer, C., Bonan, G. B. & Guenther, A. 2003 Simulating biogenic volatile organic compound emissions in the community climate system model. J. Geophys. Res. 108, 4659. (doi:10.1029/2002JD003203)
- Lihavainen, H., Kerminen, V.-M., Komppula, M., Hatakka, J., Aaltonen, V., Kulmala, M. & Viisanen, Y. 2003 Production of potential cloud condensation nuclei associated with atmospheric new-particle formation in northern Finland. J. Geophys. Res. 108, 4782. (doi:10. 1029/2003JD003887)
- Meissner, K. J., Weaver, A. J., Matthews, H. D. & Cox, P. M. 2003 The role of land-surface dynamics in glacial inception: a study with the UVic Earth system mode. *Clim. Dyn.* 21, 515–537. (doi:10.1007/s00382-003-0352-2)
- O'Dowd, C. D., Aalto, P., Hameri, K., Kulmala, M. & Hoffmann, T. 2002 Atmospheric particles from organic vapours. *Nature* 416, 497–498. (doi:10.1038/416497a)
- Randerson, J. T. et al. 2006 The impact of boreal forest fire on climate warming. Science 314, 1130–1132. (doi:10.1126/science.1132075)
- Riipinen, I. et al. 2007 Connections between atmospheric sulphuric acid and new particle formation during QUEST III–IV campaigns in Heidelberg and Hyytiälä. Atmos. Chem. Phys. 7, 1899–1914. See http://www.atmos-chem-phys.net/7/1899/2007/.
- Sampaio, G., Nobre, C., Costa, M. H., Satyamurty, P., Soares-Filho, B. S. & Cardoso, M. 2007 Regional climate change over eastern Amazonia caused by pasture and soybean cropland expansion. *Geophys. Res. Lett.* 34, L17709. (doi:10.1029/2007GL030612)
- Seinfeld, J. H. & Pandis, S. N. 1998 Atmospheric chemistry and physics: from air pollution to climate change. New York, NY: Wiley.
- Sihto, S.-L. et al. 2006 Atmospheric sulphuric acid and aerosol formation: implications from atmospheric measurements for nucleation and early growth mechanisms. Atmos. Chem. Phys. 6, 4079–4091. See http://www.atmos-chem-phys.net/6/4079/2006/.
- Spracklen, D. V., Pringle, K. J., Carslaw, K. S., Chipperfield, M. P. & Mann, G. W. 2005a A global off-line model of size-resolved aerosol microphysics: I. Model development and prediction of aerosol properties. *Atmos. Chem. Phys.* 5, 2227–2252. See http://www.atmos-chem-phys.net/5/ 2227/2005/.
- Spracklen, D. V., Pringle, K. J., Carslaw, K. S., Chipperfield, M. P. & Mann, G. W. 2005b A global off-line model of size-resolved aerosol microphysics: II. Identification of key uncertainties. *Atmos. Chem. Phys.* 5, 3233–3250. See http://www.atmos-chem-phys.net/5/3233/2005/.

- Spracklen, D. V., Carslaw, K. S., Kulmala, M., Kerminen, V.-M., Mann, G. W. & Sihto, S.-L. 2006 The contribution of boundary layer nucleation events to total particle concentrations on regional and global scales. *Atmos. Chem. Phys.* 6, 5631–5648. See http://www.atmos-chemphys.net/6/5631/2006/.
- Spracklen, D. V. et al. 2008 Contribution of particle formation to global cloud condensation nuclei concentrations. Geophys. Res. Lett. 35, L06808. (doi:10.1029/2007GL033038)
- Thomas, G. & Rowntree, P. R. 1992 The boreal forest and climate. Q. J. R. Meteorol. Soc. 118, 469–497. (doi:10.1002/qj.49711850505)
- Tunved, P. et al. 2003 One year boundary layer aerosol size distribution data from five nordic background stations. Atmos. Chem. Phys. 3, 2183–2205. See http://www.atmos-chem-phys. net/3/2183/2003/.
- Tunved, P. et al. 2006a High natural aerosol loading over boreal forests. Science 312, 261–263. (doi:10.1126/science.1123052)
- Tunved, P., Korhonen, H., Strom, J., Hansson, H. C., Lehtinen, K. E. J. & Kulmala, M. 2006b Is nucleation capable of explaining observed aerosol integral number increase during southerly transport over Scandinavia? *Tellus B* 58, 129–140. (doi:10.1111/j.1600-0889.2006.00176.x)
- Tunved, P., Ström, J., Kulmala, M., Kerminen, V.-M., Dal Maso, M., Svenningson, B., Lunder, C. & Hansson, H.-C. 2008 The natural aerosol over northern Europe and its relation to anthropogenic emissions—implications of important climate feedbacks. *Tellus B* 60, 473–484. (doi:10.1111/j.1600-0889.2008.00363.x)
- Verheggen, B., Mozurkewich, M., Caffrey, P., Frick, G., Hoppel, W. & Sullivan, W. 2007 alpha-Pinene oxidation in the presence of seed aerosol: estimates of nucleation rates, growth rates, and yield. *Environ. Sci. Technol.* **41**, 6046–6051. (doi:10.1021/es070245c)
- Zhang, Q. et al. 2007 Ubiquity and dominance of oxygenated species in organic aerosols in anthropogenically-influenced Northern Hemisphere midlatitudes. Geophys. Res. Lett. 34, L13801. (doi:10.1029/2007GL029979)
- Zhang, R. Y., Suh, I., Zhao, J., Zhang, D., Fortner, E. C., Tie, X. X., Molina, L. T. & Molina, M. J. 2004 Atmospheric new particle formation enhanced by organic acids. *Science* **304**, 1487–1490. (doi:10.1126/science.1095139)

# AUTHOR PROFILE

## **Dominick V. Spracklen**



Dominick V. Spracklen studied chemistry at the University of Leeds, graduating in 1999. He obtained his PhD in atmospheric science in 2005 before spending 2 years researching forest fires and climate at Harvard University. He is now a research fellow in the School of Earth and Environment at the University of Leeds. He has published more than 15 scientific papers on issues ranging from new particle formation to biofuels, climate change and forest fires.