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Impacts from ice-nucleating bacteria on deep convection: implications for the biosphere-atmosphere interaction in climate change

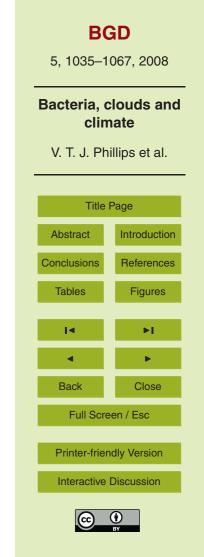
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Abstract

A cloud modeling framework is described to simulate ice nucleation by biogenic aerosol particles, as represented by airborne ice-nucleation active (INA) bacteria. It includes the empirical parameterization of heterogeneous ice nucleation. The formation of cloud

- liquid by soluble material coated on such insoluble aerosols is represented and determines their partial removal from deep convective clouds by accretion onto precipitation. Preliminary simulations are performed for a case of deep convection over Oklahoma. If present at high enough concentrations, as might occur in proximity to land sources, INA bacteria are found to influence significantly: (1) the average numbers and sizes of crystals in the clouds; (2) the horizontal cloud coverage in the free troposphere; and (3) precipitation and incident solar insolation at the surface, which influence rates of bacterial growth. At lower concentrations, the corresponding responses of cloud fields
 - appear much lower or are ambiguous.

In nature, the growth rates of INA bacteria on leaves prior to emission into the at-¹⁵ mosphere are known to be highly dependent on temperature, precipitation and plant species. Consequently, the open question emerges of whether emissions of such icenucleating biogenic particles can then be modified by their own effects on clouds and atmospheric conditions, forming a weak feedback in climate or microclimate systems.

1 Introduction

²⁰ Throughout the atmosphere, clouds are a major control on the fluxes of shortwave and longwave radiation that drive the climate system. Clouds consist of cloud particles that are either cloud droplets or crystals. When an aerosol particle (<1 μ m in size usually) "activates" at a humidity slightly above saturation, it grows very suddenly to become a cloud particle (>1 μ m). Clouds are vast sets of aerosols made visible by their activation en masse in saturated conditions. Impacts on cloud properties from changing concentrations and composition of aerosols constitute a great source of uncertainty in

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the prediction of global climate change.

Aerosols suspended in the atmosphere affect the energy budget of the Earth's climate by scattering and absorbing longwave (thermal) and shortwave (solar) radiation. They do this particularly effectively after becoming cloud particles. The concentrations

⁵ ("loadings") and chemistry of aerosols, and the rate of ascent, all determine how many of them activate to become cloud particles. This determines the average size of cloud particles, which in turn governs cloud radiative and microphysical properties, as well as cloud life-time and spatial extent (e.g. Phillips et al., 2003, 2007).

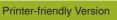
The interaction between aerosols and glaciated clouds is particularly uncertain
(Forster et al., 2007). Between 0 and -36°C (the "mixed-phase region"), liquid water can be supercooled and cloud droplets ("cloud liquid") can co-exist with crystals ("cloud ice"). Mixed-phase clouds occur at such temperatures and are important for the climate system. They influence the response of the radiation budget of the atmosphere to climate change, especially in the extratropics (e.g. Senior and Mitchell, 1993; Tsushima et al., 2006). Deep convection, very often, is partially glaciated and can be associated with horizontally extensive stratiform cloud in the mixed-phase region (e.g. nimbostratus).

In nature, there is a myriad of physical mechanisms for conversion of aerosols to cloud-particles ("heterogeneous nucleation" of cloud liquid or crystals). Soluble aerosol
 material ("cloud condensation nuclei" or CCN) can activate to become cloud droplets, as noted above. Equally, insoluble aerosol material ("ice nuclei" or IN) can nucleate ice crystals. For instance, "heterogeneous freezing" involves IN aerosols nucleating crystals when sufficiently cold (sub-zero) temperatures and high humidities (supersaturated with respect to ice) are attained. The microphysical properties and precipitation production of mixed-phase clouds are sensitive to changes of IN aerosols (e.g. Phillips et al., 2003).

Crystals may also be initiated in other ways. "Homogeneous freezing" is the type of ice nucleation that occurs spontaneously without the action of IN aerosols (e.g. at about -36° C for cloud droplets, at colder temperatures for CCN aerosols). Homogeneous

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freezing occurs at the very cold, high levels above themixed-phase region. There, clouds consist only of ice. Emission of fragments of ice during collisions in the lower half of the mixed-phase region can yield extra crystals ("ice particle multiplication"; e.g. Hallett and Mossop, 1974).

- ⁵ Diverse chemical species of aerosol in the atmosphere have always influenced climate. Primary biogenic aerosol particles (PBAs) form one category of aerosol species and are directly emitted from land sources into the troposphere. They are ubiquitous over much of the super- and sub-micron size range (e.g. Jaenicke, 2005). PBAs include pollen, bacteria, fungal spores, viruses, plant and animal fragments (e.g. Despres et al.,
- 2007). PBAs co-exist in the atmosphere with many other species of aerosol (e.g. soluble sulphate and seasalt particles, insoluble dust and soot particles). If PBAs become coated with soluble material during atmospheric transport (or if their surface is wetable), they can act as CCN and become activated as cloud droplets (e.g. Möhler et al., 2007). Because every PBA has an insoluble core, PBAs can sometimes act as IN and nucleate ice.

Analyses of the residual material in ice crystals nucleated heterogeneously in the atmosphere have revealed that most are formed on IN of dust and carbonaceous aerosol (e.g. De- Mott et al., 2003a). A fraction of these carbonaceous insoluble aerosols are organic (e.g. Targino et al., 2007) and may be biogenic (e.g. PBAs). Their precise type of biological origin is not yet clear. Laboratory experiments have shown that certain ice-nucleation active (INA) bacteria (e.g. Vali et al., 1976; Lindow et al., 1978), leaf litter (Schnell and Vali 1972, 1976), pollen (Diehl et al., 2001; Diehl and Wurzler, 2004) and a solid hydrate of oxalic acid (Zobrist et al., 2006) can all nucleate ice heterogeneously. In freshly fallen snow, ice nucleators of biological origin active at relatively warm subzero temperatures (between -2 and -7°C) are present at concentrations of up to 120 particles per litre of melted snow (Christner et al., 2008). The relative extents to which these substances actually do nucleate ice in the atmosphere is still a moot

Unequivocally, meteorological conditions near the surface determine the land

point (e.g. Möhler et al., 2007).

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sources of such PBAs (e.g. INA bacteria) in several ways (Sect. 2). But conversely, there is the open question of whether emissions of biogenic IN from sources at the ground can significantly modify cloud properties. If so, a feedback between their growth, emissions and the local (micro-) climate then emerges. For instance, additional

⁵ crystals from airborne INA bacteria might alter the radiative properties of clouds, as investigated here. Similarly, a two-way relationship between the rain and bacteria in the biosphere was hypothesized by Sands et al. (1982). INA bacteria have been seen to follow closely the water cycle of precipitation (Sands et al., 1982; Morris et al., 2008a). Biological ice nucleation may be viewed as a potential aspect of the aerosol/ice-cloud interaction in climate change.

Cloud processes are represented by "bulk microphysics schemes" within cloudsystem resolving models (CSRMs). Some bulk schemes predict the numbers of cloud droplets and crystals as advected quantities, allowing their sizes to be predicted. Such schemes are referred to as "double-moment". They represent the diversity of mechanisms by which cloud particles are formed from aerosol particles (e.g. Phillips et al., 2007).

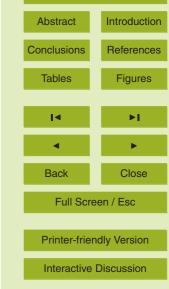
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The aim of the present paper is to assess qualitatively the potential impacts of airborne ice-nucleating bacteria on cloud properties and how conditions near the surface are then modified. An advanced CSRM with a double-moment bulk representation of clouds is applied. We have included in the CSRM a representation of biological nucleation of crystals and cloud droplets. The cloud model and simulated case are

- described in the next section. Subsequent sections show results for simulated scenarios with various atmospheric loadings of INA bacteria for an observed summer-time continental case of deep convection. The location, Oklahoma, is where high loadings
- of airborne INA bacteria would be expected downwind from cultivated fields and other ground sources in early summer. In the final section, implications for the atmospherebiosphere interaction are discussed. Focus is given to the potential existence of some kind of (micro-) climate feedback involving the growth and atmospheric nucleation of INA bacteria.

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2 Origins of ice-nucleating bacteria

Advanced measurement techniques, such as epifluourescent spectroscopy (Harrison et al., 2005), now reveal total concentrations of all bacterial cells in the free troposphere that are highly variable $(10-1000 L^{-1})$, as reviewed by Phillips et al. (2008).

⁵ The vast majority of these airborne cells are non-culturable ("non-viable") (e.g. Bauer et al., 2002), partly owing to dessication and exposure to ultra-violet light. Previous estimates were much lower and were from counting only culturable ("viable") bacterial cells, which form a very tiny minority (about 0.1%).

A few of the airborne bacteria are known to nucleate ice. A commonly occurring species of bacteria, Pseudomonas (P.) syringae, that grows on plant surfaces has been seen to have ice-nucleating properties (e.g. Vali et al., 1976; Lindow et al., 1978; Lindemann et al., 1982; Gross et al., 1983; Hirano et al., 1985). In the Mid-West of the USA (North and South Dakota, Wisconsin, Kansas) P. syringae has been a plant disease afflicting fields of wheat and barley. It was rife during the 1970s. Cells of P. syringae can

- reach high numbers on plants in the cool early growing season. These INA bacteria can be dispersed into the atmosphere from leaves. Diverse strains have a spectrum of varying degrees of nucleating ability, with freezing fractions spanning about six orders of magnitude (e.g. Hirano et al., 1985). Furthermore, there is biological evidence that P. syringae closely follows the water cycle (Sands et al., 1982; Morris et al., 2008a).
- The same clone isolated from disease epidemics of a crop plant, from lakes used for irrigation and from freshly fallen snow, illustrates the cycling of this bacterium between land sources and the atmosphere. Salient features of INA bacteria in general are summarised by Morris et al. (2004).

Most of the micro-organisms known to be ice nucleators are plant-associated. These include the bacteria Xanthomonas sp., Pantoea agglomerans, and other Pseudomonas spp. as well as the plant pathogenic fungus, Fusarium (F.) avenaceum (as reviewed by Morris et al., 2004). Interestingly, in addition to P. syringae, Amato et al. (2007) have reported the isolation of F. avenaceum from clouds at about 1450 m



altitude in central France.

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Climate determines the land sources of INA bacteria in several ways. Firstly, observations of bacterial growth, including that of P. syringae in particular, were encapsulated in a model by Caristi et al. (1991). It predicted that the growth of P. syringae populations on leaf surfaces depends on the length of time a leaf remains wet after rain

- populations on leaf surfaces depends on the length of time a leaf remains wet after rain or dew (the drying period). Secondly, only within a certain temperature range (e.g. between 2 and 32°C; Caristi et al., 1991) is growth of the bacterial population observed. Its rate depends exponentially on temperature, and order-of-magnitude variations in cell concentration are seen to follow the diurnal cycle of surface air temperature. Out-
- side this range of temperature, the bacteria are either in a sort of state of suspended animation known as hypobiosis (Linton, 1971) or may be killed altogether at warm temperatures. Lindow et al. (1978) observed that concentrations of culturable INA bacteria on leaves of crops increased exponentially with time during the summer in Wisconsin, by up to about 4 orders of magnitude. INA bacteria were about 1% of the total number
- of bacteria, but this fraction was also found to be variable (Lindemann et al., 1982). Average concentrations of airborne bacteria were seen to increase exponentially with temperature in the UK by Harrison et al. (2005). Order-of-magnitude variations between all four seasons were seen in relative contributions from various genera, such as Pseudomonas. Finally, wind speed modulates bacterial concentrations (Harrison et al., 2005).

Biological ice nucleators in freshly fallen snow were more abundant by about 50fold in snow collected from inland regions of the US and Europe compared to snow collected in Antarctica (Christner et al., 2008). This offers futher support for the overall importance of vegetated land masses as the principle origin of bacterial and other biological ice nucleators in the atmosphere.

By contrast, bacteria and viruses isolated from aquatic environments near sea-ice in polar regions display an unusual absence of ice-nucleating activity at the temperatures to which sea-water may be supercooled (Junge and Swanson, 2007). If these isolates are representative of the majority of micro-organisms in such polar environments, then

it suggests that avoidance of ice formation may be one of the coldadaptation and survival strategies for sea-ice bacteria. That extreme example illustrates how generally, on Earth, certain micro-organismsmay have evolved an enhanced or suppressed icenucleating ability according to whether it confers an advantage for survival in any given niche. Indeed, the capability for survival of bacteria in glaciated environments must be

a crucial aspect of biosphere systems (e.g. Junge and Swanson, 2007). Much of the Earth's surface is periodically covered by ice during the glacial cycle over geological time yet life-forms have successfully adapted to such adverse events nonetheless.

Air-borne dissemination is a key stage of the life cycle of many micro-organisms, par tially determining their geographical distribution. In the evolution of micro-organisms such as INA bacteria, adaptation to atmospheric conditions probably influenced their genome structure (Morris et al., 2008b). For instance, nucleation of cloud particles by (e.g. INA) bacteria provides a clear mechanism for their return to the Earth's surface. The nucleating ability of bacteria might conceivably have evolved in response to selection pressures. Such selection, related to the interplay of micro-organisms with atmospheric conditions, is being explored in the emerging field of bio-meteorology.

3 Description of cloud model

3.1 Overview

The cloud model applied in the present study is a CSRM developed by Phillips and ²⁰ Donner (2007) from a version of the Weather, Research and Forecasting (WRF) model. Double-moment bulk microphysics with interactive radiation was incorporated by Phillips et al. (2007). More recently, the CSRM has been upgraded to include a fully interactive component of aerosol (a "prognostic aerosol component"). It now represents depletion of environmental aerosols as they activate to become cloud droplets that are

then removed by accretion onto precipitation ("in-cloud nucleation scavenging"). A novel empirical parametrisation by Phillips et al. (2008) predicts crystal nucleation by





In nature, if such hygroscopic, soluble coatings are deposited onto bacteria before or after emission, their uptake of liquid water would reduce the cells' dessication. Leaf surfaces are subjected to cyclic periods of drying. Such a mechanism for water uptake

insoluble organic aerosols, such as INA bacteria and other biogenic particles.

3.2 Coupled aerosol and cloud components

The bulk microphysics parameterization (Phillips et al., 2007) has 5 classes of hydrometeor (cloud liquid, cloud ice, snow, graupel and rain), and includes sedimentation of cloud ice and cloud liquid. The scheme is double-moment insofar as the numbers of cloud droplets and crystals are predicted as well as their mass. No saturation adjustment or parametrisation of the Bergeron-Findeisen process is applied, since diffusional growth of cloud particles and precipitation is predicted explicitly with a linearised supersaturation scheme.

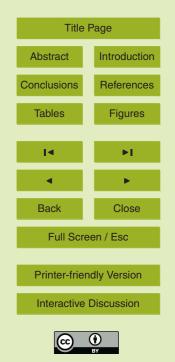
¹⁰ The predicted supersaturation determines nucleation of cloud particles, coupling the cloud and aerosol components in the CSRM. The prognostic aerosol component includes three soluble and three insoluble chemical species (sulphate, seasalt and soluble organics; dust, soot and insoluble organics). Heterogeneous nucleation of crystals and droplets by all six species is represented with the empirical parametrisation by

- Phillips et al. (2008) and a scheme by Ming et al. (2006). In-cloud nucleation scavenging of all aerosol species is represented, with their separate concentrations in air, cloud particles and precipitation being explicitly resolved. In-cloud scavenging of insoluble aerosols (e.g. biogenic particles) is treated by assuming each particle has been coated with soluble material (e.g. by atmospheric chemical processing), with an as-
- ²⁰ sumed fraction of its total mass being soluble (Clarke et al., 2004). Köhler theory (Petters and Kreidenweiss, 2007) determines whether an insoluble particle's coating of soluble material activates as a cloud droplet, immersing it. CCN activity of biogenic aerosol particles is related to their coatings of soluble material, providing a source of extra droplets. The empirical parametrisation also represents heterogeneous freezing of rain by the insoluble aerosol acquired by accretion of cloud liquid.

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would provide a potential mechanism for enhanced survival and growth of bacteria.

Known mechanisms for nucleation of cloud particles are represented, as well as the Hallett-Mossop process (Hallett and Mossop, 1974) of ice particle multiplication. Homogeneous freezing of supercooled cloud liquid allows for evaporation of a fraction

of its mass without freezing and occurs at the -36°C level. Homogeneous aerosol freezing is represented at much colder levels. Droplet nucleation occurs at cloud base, but also sometimes in the interior of the cloud when the supersaturation becomes high enough.

Autoconversion of cloud liquid to rain depends on the mean droplet size implied by the predicted droplet number (and similarly for the autoconversion of cloud ice to snow) and on the width of the droplet size distribution. Conversion of snow to graupel by riming has been expressed in terms of the riming rate. Accretion of particles of cloud liquid and cloud ice represents the dependence of collision efficiency on their size.

3.3 Biological ice nucleation

- An empirical parameterization of heterogeneous ice nucleation by Phillips et al. (2008) has been implemented in the CSRM, as noted above. It is based on coincident field measurements of IN activity and aerosol loadings in the free troposphere on Mt Werner in Colorado (DeMott et al. 2003a). It includes dependencies on the predicted (surface area) mixing ratio of three groups of chemical species of IN aerosols. One of the groups consists of insoluble organic (e.g. biogenic) aerosols. The empirical parameterization
- ²⁰ consists of insoluble organic (e.g. biogenic) aerosols. The empirical parameterization represents all knownmodes of heterogeneous nucleation (condensation-, immersion-and conventional/inside-out contact-freezing modes, in addition to vapour deposition). As an input, it requires prediction by the model of the supersaturation at the cloud scale.
- The empirical parameterization enables a representation of biological ice nucleation. INA strains of bacteria have been selected as partially representative of the group of insoluble organic IN aerosols. About half of the crystals from this group are estimated to originate from INA bacteria, though this estimate is uncertain. Cells belonging to



INA strains form only a small fraction (1%; Lindemann et al. 1982) of all non-viable and viable airborne bacteria cells (10–1000 L⁻¹) in the troposphere (Sect. 2), which are estimated to have an average concentration of about 100L⁻¹. Observed fractions of the INA bacteria that freeze (e.g. Vali et al., 1976; Gross et al., 1983; Hirano et al., 1985) during supercooling partly constrain the treatment of insoluble organic IN (e.g. INA bacteria) for the empirical parameterization by Phillips et al. Insoluble organic IN are supposed to nucleate about 6% of all heterogeneous crystals in the background free troposphere. But there is significant uncertainty associated with this estimate fraction, owing to ambiguity in analysis of the composition of IN inside heterogeneously
nucleated crystals sampled from the atmosphere and due to uncertainty in total cell concentrations.

The empirical parameterization predicts that the contribution frominsoluble organic aerosols varies with their loading. Figure 1 shows that the predicted freezing fraction for insoluble organic IN from the empirical parameterization (Phillips et al., 2008) agrees with independent loboratory observations of INA bestaria (platted in blue). Timical

- ¹⁵ with independent laboratory observations of INA bacteria (plotted in blue). Typical observed values of mean aerosol size and spectral width were applied (Phillips et al., 2008) to infer the predicted freezing fraction, which is robust for a wide range of aerosol scenarios and does not depend on any assumed aerosol concentration. The observed freezing fractions for single INA strains vary by many orders of magnitude at any given
- ²⁰ supercooling, as expected from the lognormal distribution of nucleating ability among strains seen by Hirano et al.

4 Experimental design

periodic for all prognostic variables.

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In these experiments, the CSRM is integrated with horizontal and vertical resolutions of 2 and 0.5 km. The vertical grid has 40 levels. A time step of 10 s is used. The domain is 2-D and has a width of 170 km (85 grid points). All lateral boundary conditions are



4.1 Control run

A control simulation of an observed case of deep convection from Oklahoma in the summer of 1997 has been performed from the Atmospheric Radiation and Measurement campaign (ARM, Sub-case C, 13–17 July 1997) in the manner described by

- ⁵ Phillips and Donner (2007). The CSRM is initialised by observed profiles of temperature and humidity (Zhang et al., 2001), and with profiles of aerosol species predicted by the GFDL GCM (GFDL GAMDT, 2004; Horowitz, 2006). Convection is maintained by applying tendencies to represent large-scale advection of humidity and temperature from Zhang et al. into the simulated domain. Perturbations are imposed onto the ini-
- tial field of vapour mixing ratio so as to initiate convection. Observed fluxes of heat and moisture, and other observed properties (e.g., ground temperature), of the land surface are prescribed. The ground temperature is not predicted yet, owing to the absence of an interactive land component. Environmental aerosol species are replenished by evaporation of cloud- and precipitation in the atmosphere, and are depleted by
- ¹⁵ nucleation. Aerosol profiles are restored to initial values once every 24 hours, following Salzmann et al. (2007). A comparison with independent (e.g., satellite) observations, of an earlier version of the CSRM without coupling of cloud and aerosol components, is documented by Phillips and Donner (2007).

For the current control run, a total concentration of all airborne bacterial cells (INA and non-INA, viable and nonviable) of about 100L⁻¹ has been assumed for the background free troposphere (Sect. 3.3; Phillips et al., 2008). A fraction (1%) are assumed to be INA bacteria.

4.2 Zero, high and ultra-high bacterial cases

Throughout the global troposphere the total concentration of all airborne bacterial cells is highly variable, being in the range of about 10–1000 L⁻¹ (Sect. 3.3). It is conceivable that there might be total concentrations of all bacteria even higher than 1000 L⁻¹ just downwind from nearby land sources of INA bacteria (e.g. cultivated fields). Ob-

servations with modern, accurate techniques (e.g. epifluorescent microscopy; Sect. 2) of all airborne bacterial cells in proximity to their sources are scarce. Conventional techniques for counting cells by growing them on plates neglect the vast majority of airborne cells that are non-culturable in the atmosphere (Sect. 2).

- ⁵ Consequently, the environmental loading of insoluble organic aerosol in the background troposphere is increased by factors of 10 and 100 in perturbation simulations (the "high and ultra-high bacterial cases" respectively), relative to the control. They explore scenarios with total bacterial concentrations that are higher by similar factors. An extra run is performed with all insoluble organic aerosol excluded from the control run (the "zero bacterial case"). In all other respects, these simulations are identical to
- the control run.

The perturbation simulations are idealised in the sense that the modified loadings of organic aerosol do not reflect any variability of biogenic aerosol actually seen over Oklahoma itself. No field experiments have been done there to provide such information.

- We selected the ARM case in Oklahoma because it uniquely includes observations of largescale advection of heat and moisture needed to force the model during multi-day simulations. Nevertheless, generally in the Mid-West USA (North and South Dakota, Wisconsin, Kansas) P. syringae has been observed to be rife as a plant disease in fields of wheat and barley (e.g. during the 1970s). Cells of P. syringae were typically
 seen to reach high numbers in the cool early growing season. Consequently, such scenarios of conjous airborne INA bacteria are plausible for Oklahoma in early sum-
- scenarios of copious airborne INA bacteria are plausible for Oklahoma in early summer near sources on the ground.

5 Results from CSRM simulations: role of ice-nucleating bacteria

Sensitivity studies are shown of the impact that biogenic aerosols could have on cloud properties for a realistic range of scenarios of bacterial concentrations in simulations with the CSRM (Sect. 3) of the ARM case (Sect. 4). The zero (×0), high (×10) and ultra-high (×100) bacterial cases defined above are compared with the control run.

5.1 Impacts on properties of visible cloud:

Figure 2 shows that when upper-level cirrus is absent, an approximate doubling and tripling of the average number concentration of crystals in visible cloud is predicted in the high and ultra-high bacterial cases respectively, relative to the control. However,
throughout the rest of the simulated period, such modification of heterogeneous ice nucleation has little impact on crystal numbers because then the convection is deeper and dominated by homogeneous freezing aloft.

Figure 3 shows corresponding perturbations of the cloud droplet concentrations associated with the CCN activity of biogenic particles and their nucleation of crystals. Little systematic, significant change in droplet concentration is evident.

Figure 4 shows a reduction by up to $10\,\mu$ m (about 10%) in average crystal size at most cloud levels in the mixed-phase region (about 4–9 km altitude above ground; 0 to -36° C) when upper-level cirrus is absent in the high and ultra-high bacterial cases relative to the control. A similar reduction (10–50 μ m) is found in the control simulation

relative to the zero bacterial case. Such results are consistent with intensified competition between more crystals for available vapour during diffusional growth of crystals in widespread cloudy regions of weak ascent. There the supercooled cloud liquid can easily vanish by evaporation (e.g. Phillips et al., 2007).

Figure 5 shows significant perturbations of the mean cloud-droplet diameter (by up to 5 μ m), caused by corresponding perturbations in the cloud-droplet concentration noted above. Nonlinearity of the microphysics-dynamics system explains the complexity of the responses of cloud liquid properties to increased crystal concentrations (e.g. by alteration of supersaturations or riming rates). Such changes would be expected to affect the generation of precipitation (e.g. by the warm rain process).

²⁵ 5.2 Impacts on meso-scale atmospheric conditions:

Figure 6 shows that the cloud fraction is higher at most cloudy levels in the ultra-high bacterial case relative to the control. The enhancement of cloud cover is especially

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marked at levels in the mixed-phase region (about 4–9 km altitude above ground) where heterogeneous ice nucleation and H-M ice multiplication (about 5 km altitude) occur. The responses of cloud fraction in the zero and high bacterial cases are ambiguous and minimal respectively.

- ⁵ Figure 7 shows that the downward component of the shortwave flux near the ground (averaged over the whole domain) is reduced by over 10 W m⁻² in the ultra-high bacterial case, owing to more reflection of incoming solar radiation by clouds that are more extensive and reflective, due to more numerous and smaller crystals. This type of impact on cloud radiative properties and domain-wide radiative fluxes can be caused by machine the machine the machine terms of any appeale of an
- ¹⁰ by modified loadings of any species of aerosol, not only of insoluble organic aerosol. Figure 8 shows that the corresponding change in the downward longwave flux is an increase by about 2 W m⁻². Clearly, there appears to be a threshold behaviour of cloud radiative properties with respect to increases in the insoluble organic aerosol loading.

Figure 9 shows that the accumulated surface precipitation is suppressed slightly by ¹⁵ up to 2 mm (about 10%) in the ultra-high bacterial case relative to the control, during most of the precipitating period of the simulation. There is little change in precipitation in the other perturbation simulations that involve less change of the atmospheric loadings of insoluble organic aerosol.

6 Conclusions

A framework has been described for modeling the effects of biogenic particles on clouds. It utilises the empirical parameterization of heterogeneous ice nucleation of multiple chemical species of aerosol (Phillips et al., 2007). A case of deep convection over Oklahoma has been simulated. Elevated loadings of biogenic aerosol, as might occur downwind from land sources, are represented by boosting the environmental concentration of insoluble organic aerosol.

The main conclusions are summarised as follows:

- The average concentration of crystals is increased by up to about half an order of magnitude in the ultrahigh bacterial case within cloudy regions without upper level-cirrus relative to the control. Sub-saturation from extra crystals' vapour growth (e.g. Phillips et al., 2007) must limit ice nucleation and multiplication in cloudy regions of weak ascent, quenching some of the sensitivity.

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- Slight perturbations in the properties of cloud-liquid (e.g. mean droplet size, droplet concentration) are found in the ultra-high bacterial case relative to the control. They arise from the altered glaciation and from the CCN activity of the extra biogenic particles. Such changes must perturb the warm rain process.
- The downward shortwave flux is reduced appreciably (by about 10 W m⁻² or 5%) in the ultra-high bacterial case relative to the control. This arises from extra reflection of incoming solar radiation by clouds that are more extensive and have more numerous and smaller crystals, which enhances their albedo. The shortwave reduction is only slightly counteracted by a weak increase (2 W m⁻²) in the downward longwave emission to the surface. If sustained over a period of weeks, such flux changes would tend to cool the land surface.
 - Surface precipitation is slightly reduced (by about 10% temporarily) in the ultrahigh bacterial case relative to the control. This may be explicable in terms of boosted evaporation of rain that must fall from higher levels when the vapour growth of more numerous crystals reduces the humidity, reducing the sizes of the largest droplets in the rain-generating parts of cloud. The CCN activation of the biogenic particles to form cloud droplets is represented in the model and may also affect the warm rain process (coalescence of droplets to form raindrops). Since the convection is deep and vigorous, the warm rain process is the dominant mechanism for generation of surface precipitation.
 - These responses of cloud cover and properties display a threshold behaviour insofar as they are ambigous or minimal in the zero and high bacterial cases



relative to the control, but suddenly become substantial in the ultra-high bacterial case. A threshold response to increasing IN concentrations in vigorous deep convection was found by Phillips et al. (2007) also for tropical oceanic cases, with a major response occurring at extremely high concentrations of IN and little impact otherwise.

The insoluble organic aerosol in the ultra-high bacterial scenario boosts by only a factor of about six the total number of active IN fromall aerosol groups (dust, soot, biogenic) relative to the background troposphere. This is much less than the boosting of IN concentrations commonly seen from common emissions of dust (e.g. DeMott et al., 2003b; Phillips et al., 2007) or soot. It partially explains why the impacts found here, on clouds from biological ice nucleation, are quite modest.

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Nevertheless, these results show that emissions of INA bacteria into the atmosphere, if sufficiently intense, can affect the meteorological conditions (surface wetness and temperature) determining their own rates of growth on leaves emerges. This connotes

the potential existence of a weak (micro-) climate feedback of some kind, similar to that hypothesized by Sands et al. (1982). Growth rates of INA bacteria on leaves (Sect. 2) are known to vary with: – (1) surface air temperature, exponentially; and (2) the drying period after precipitation has fallen at the ground. In nature, changes in the intensity of precipitation and in fluxes of incident solar radiation at the surface, as simulated in the present study, would be expected to modify both factors.

Yet there is much uncertainty in the empirical parameterization's representation of biological ice nucleation (Sect. 3.3; Phillips et al., 2008). The order of magnitude of the total concentration of all cells (viable and non-viable, INA and non-INA) in the back-ground troposphere is uncertain, because only recently have advanced measurement

techniques been available to count all of them (e.g. microscopy). Also, the fraction of heterogeneous crystals nucleated by INA bacteria has not yet been observed directly, owing to limitations in the chemical analysis. In view of these possible biases of the cloud model, the results documented here may be viewed qualitatively.

The vigour and depth of convection in the ARM case tended to obscure atmospheric



effects from bacterial ice nucleation in the present study. Vigorous deep convection can reach the very cold temperatures aloft (e.g. above about 9 km altitude) required for homogeneous freezing (no action of IN). Generally, homogeneous freezing dominates overall ice nucleation whenever it can occur (e.g. Phillips et al., 2007). Changes

in insoluble aerosol loadings are found here to have a more discernable effect when the convection is not deep enough to extend above the mixed-phase region where homogeneous freezing would occur. This is why light precipitation from mixed-phase nimbostratus is highly sensitive to IN concentrations (e.g. Phillips et al., 2003). Such horizontally extensive cloud is ubiquitous at mid- to high-latitudes, where INA bacteria are common.

Future work may focus on simulation of such clouds that are less deep. An ARM case could be artificially modified (e.g. by imposing a temperature inversion below the level of upper-level cirrus), or a stratiform case simulated. Inclusion of an interactive land component would allow the ground temperature to be predicted, so the response of air temperature near the surface could be assessed. Ensembles of simulations, or a

larger (e.g. 3-D) domain, would increase the statistical significance of results. Biogenic particles can act as CCN and the impact from their IN activity can be amplified by the H-M process of ice particle multiplication (-3 to -8°C) (e.g. Möhler et al.,2007). Such processes are treated by our cloud model with its inclusion of in-cloud

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- scavenging (Sect. 3) and their roles may be analysed in future work. The potential for super-micron biogenic particles to accelerate coalescence by acting as giant CCN, creating rapidly falling cloud droplets early during ascent, is not explicitly represented with the autoconversion scheme currently applied in the CSRM. However, the role of giant CCN has been found to be minimal when clouds are far from the threshold for
- the onset of warm rain (Feingold et al. 1999), which is true of deep convection. By contrast, in-cloud turbulence intensifies rain formation in deep convection appreciably (e.g. Phillips et al., 2005).

Finally, a novel framework has been proposed for modeling biological ice nucleation and the biosphere-atmosphere interaction. The modeling framework is apt for inclu-



sion of a component treating the growth of INA bacteria on leaves in the biosphere (e.g. Caristi et al., 1991). In future, more extensive, accurate and correlated field measurements of the occurrence and nucleating ability of multiple species of biogenic IN may be easily assimilated into the framework, improving model accuracy. In particular,

⁵ fresh analysis of the composition of biogenic residual particles at the centres of heterogeneously nucleated crystals (e.g. Cziczo et al., 2004) may permit a more detailed treatment of biological ice nucleation by the empirical parameterization. Field measurements of INA bacteria and coincident meteorological observationsmay provide in future better cases for simulation with advanced cloud models. An inter-disciplinary focus will be needed (Morris et al., 2008b).

In summary, airborne INA bacteria, if present at sufficiently high concentrations, are shown here to modify significantly the solar insolation and precipitation near the ground. These are quantities known to determine bacterial growth. The potential existence of a weak (micro-) climate feedback between biological ice nucleation, clouds and emissions of biogenic particles merits further exploration.

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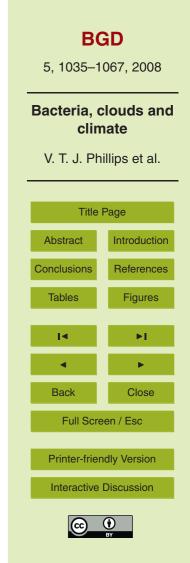
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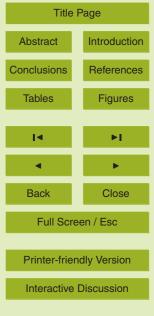
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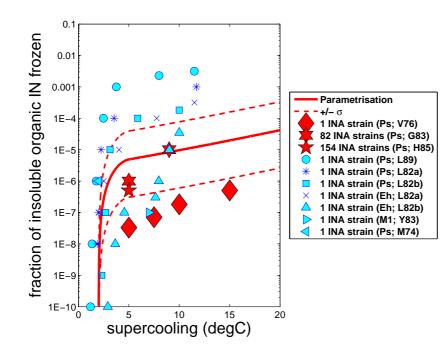


Fig. 1. Frozen fraction of insoluble organic particles predicted by the empirical parametrisation and inferred from laboratory studies of the immersion-freezing of bacteria. The observed freezing fractions for INA strains have been multiplied by 0.01 (Lindemannn et al., 1982) to convert them to freezing fractions for all strains (INA and non-INA). Many INA strains of Pseudomonas syringae ("Ps") were analysed by Gross et al. (1983 ['G83']) and Hirano et al. (1985 ["H85"]). The freezing of single INA strains of Ps (Maki et al., 1974 ["M74"]; Vali et al., 1976 ["V76"]; Lindow, 1982 ["L82a"]; Lindow et al., 1982 ["L82b"]; Lindow et al., 1989 ["L89"]), M1 (Levin and Yankofsky, 1983 ["LY83"]) and Erwinia Herbicola ("Eh"; L82a,b) are also shown. Except for observations by Gross et al., Hirano et al. and Vali et al. (red symbols), the displayed data were not used in construction of the scheme.



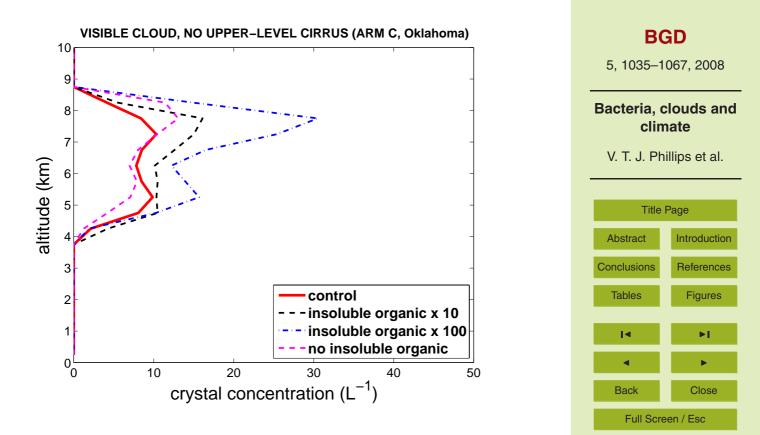
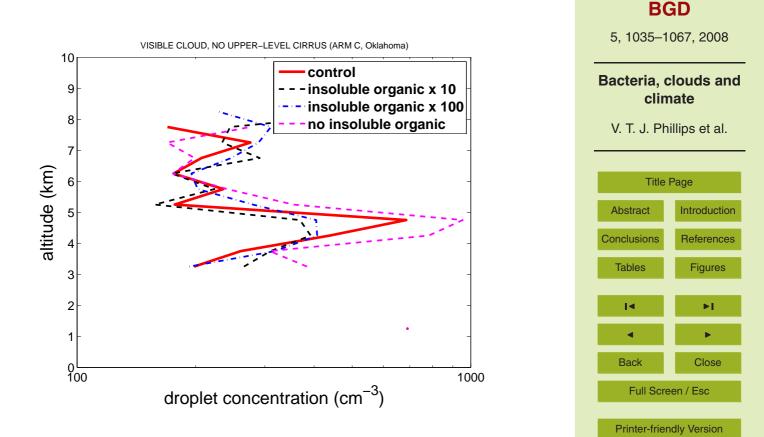
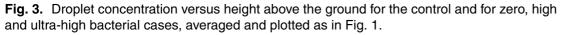


Fig. 2. Crystal concentration versus height above the ground for the control and for zero, high and ultra-high bacterial cases, conditionally averaged over visible cloudy regions without visible upper-level cirrus. Any grid-point is deemed to be cloudy and visible when its normal optical depth exceeds an assumed threshold (0.2).

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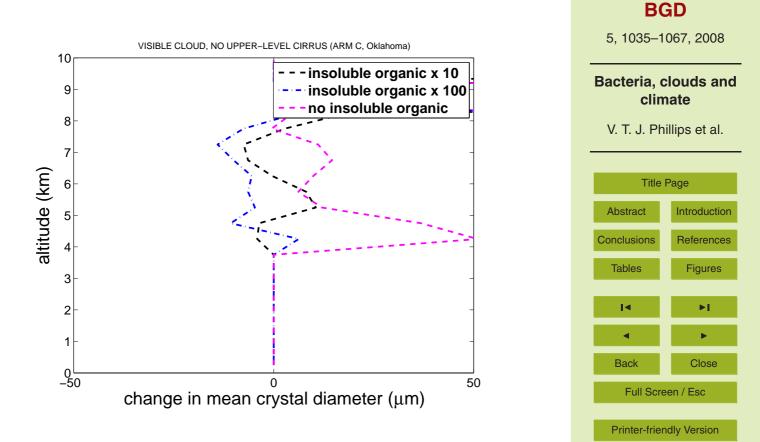


Fig. 4. Change in average profiles of mean crystal size versus height above the ground for zero, high and ultra-high bacterial cases relative to the control, with averaging as in Fig. 1.



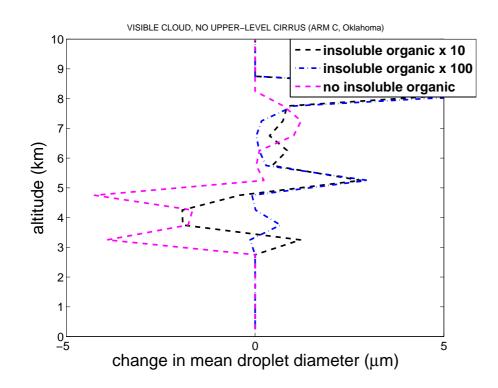
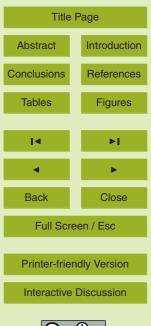


Fig. 5. Change in average profiles of mean droplet size versus height above the ground for zero, high and ultra-high bacterial cases relative to the control, with averaging as in Fig. 1.

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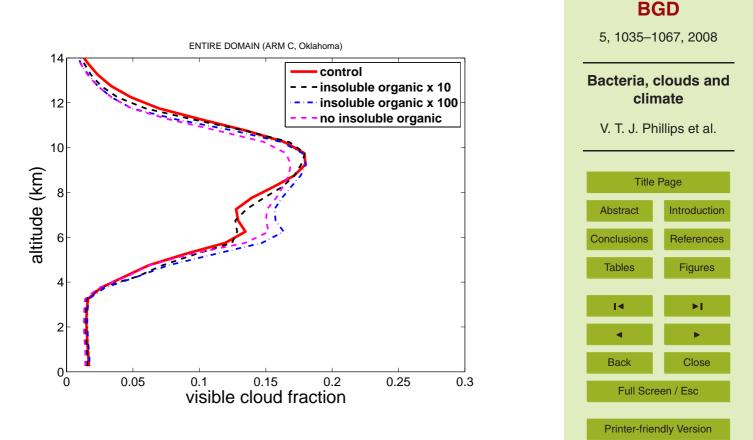


Fig. 6. Average fraction of meso-scale domain covered by visible cloud (defined as in Fig. 1) in the control, zero, high and ultra-high bacterial cases, versus height above the ground.



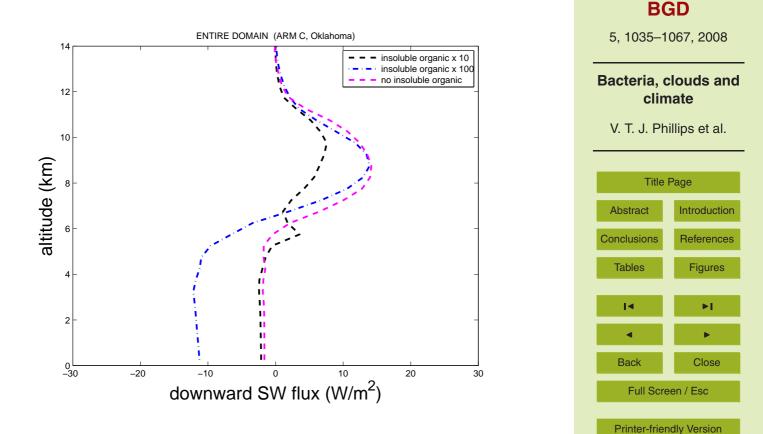


Fig. 7. Change in domain-averaged profiles of the downward component of the shortwave flux versus height above the ground for zero, high and ultra-high bacterial cases relative to the control.



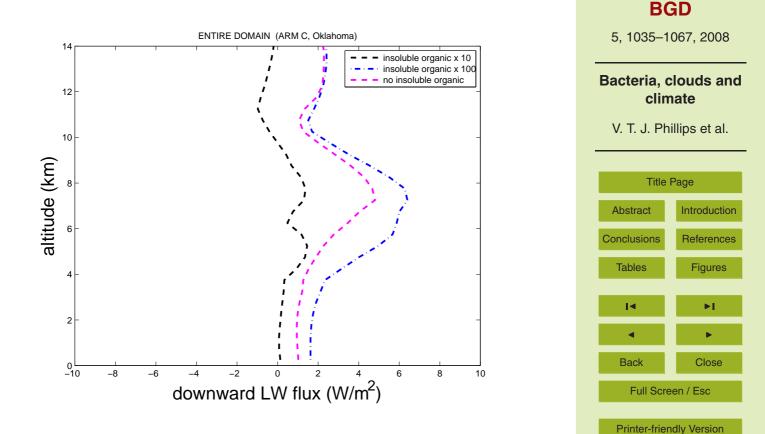


Fig. 8. Change in domain-averaged profiles of the downward component of the longwave flux versus height above the ground for zero, high and ultra-high bacterial cases relative to the control.

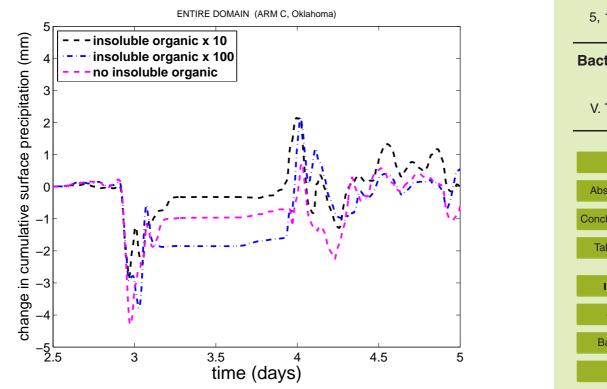


Fig. 9. Change in accumulated surface precipitation (mm) for zero, high and ultra-high bacterial cases relative to the control, as time progresses (days after start of simulation). The first two days of the simulation are omitted here as no significant precipitation occurred then.

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