Cloud effects from boreal forest fire smoke: evidence for ice nucleation from polarization lidar data and cloud model simulations

Kenneth Sassen\(^1\) and Vitaly I Khvorostyanov\(^2\)

\(^1\) Geophysical Institute, University of Alaska Fairbanks, 903 Koyukuk Drive, Fairbanks, AK 99775, USA
\(^2\) Central Aerological Observatory, Moscow, Russia

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Abstract
Polarization lidar observations from the interior of Alaska have revealed unusual supercooled altocumulus cloud conditions in the presence of boreal forest fire smoke from local and regional fires. At temperatures of about \(-15\) °C, the lidar data show ice nucleation prior to liquid cloud formation (i.e. below water saturation), as well as the occasional glaciation of the liquid layer. Thus the smoke aerosol appears to act as ice nuclei that become activated in updrafts before the liquid cloud forms, as the concentrated aqueous organic solutions are diluted sufficiently to allow them to freeze heterogeneously. This haze particle freezing process is similar to the production of cirrus ice crystals homogeneously at much colder temperatures. To test this hypothesis, cloud microphysical model simulations constrained by the measurements were performed. They indicate that this heterogeneous ice nucleation scenario can be supported by the cloud model. Although ice formation in this manner may generally act in the atmosphere, the boreal smoke particles produce an unusually dramatic effect in the lidar data. We conclude that smoke-induced ice nucleation occurs at moderate supercooled temperatures either through the effects of raised soil/dust particles embedded in the smoke droplets, coated soot aerosol or through the nucleation via certain organic solutions.

Keywords: indirect aerosol/cloud effects, boreal smoke, polarization lidar

1. Introduction
One possible response to global warming is an increased risk of forest, brush, grassland and tundra fires across the globe. Forest fire activity appears to be particularly susceptible to changes in the seasonality of precipitation, the onset of snowmelt and drought stress, all factors that could accompany regional climate change. Moreover, the increased carbon flux to the atmosphere during burning could exacerbate global warming as a positive feedback. The direct radiative effects of the aerosol burden from biomass burning (both natural and anthropogenic) can significantly affect the atmospheric radiation balance (Kasischke and Penner 2004), while the magnitude of the indirect effects on clouds remains largely a mystery. Possible indirect effects on clouds include smoke aerosol-induced changes in cloud droplet size spectra, effects on cloud thermodynamic phase and even on the characteristics of lightning in convective clouds (Fernandes et al 2006). Such changes in clouds come about when smoke particles compete successfully with other aerosols as cloud condensation nuclei (CCN) or ice nuclei (IN), changing the cloud contents and resulting in alterations in radiative transfer and potentially our climate. For example, modeling studies of supercooled altocumulus clouds found significant differences in the longwave and shortwave radiation fluxes through the atmosphere, and in heating rates, as a water cloud gradually glaciated into an ice cloud (Sassen and Khvorostyanov 2007).
It appears that the effects of climate change are especially noticeable in polar regions because of the action of various feedback mechanisms and, indeed, a recent increasing trend in North American boreal forest burning has been documented (Kasischke and Turetsky 2006). In Alaska, two recent fire seasons have been among the most severe in terms of total areas burned since 1955, when records were started by the Alaska Forest Service. During the summer of 2004 a record 26 166 km$^2$ were burned and in 2005 a total of 18 616 km$^2$ burned, which represents the third-most devastating fire season on record. The aerosol produced by the fires was regularly observed in the interior of Alaska by a polarization lidar at the University of Alaska Fairbanks. Among remote sensors, polarization lidars have shown a unique sensitivity to particle shape, which leads to the unambiguous discrimination between liquid and solid phase cloud particles and the identification of major aerosol types (Sassen 2000, 2005, 2008). For example, non-spherical desert dust aerosols produce relatively strong laser light depolarization, while spherical haze or smoke particles produce very little or none (Murayama et al 2004).

Below, we begin by reviewing the utility of polarization lidars for studying clouds, aerosols and their interactions.

2. Polarization lidar aerosol research capabilities

A common form of the lidar equation describing the backscattered power $P$ received as a function of range $R$ is given by

$$P(R)_{∥,∥} = P_o K \beta(R)_{∥,∥} \exp \left[-2 \int \sigma(R) dR \right] / R^2$$

(1)

where $P_o$ is the power out, $K$ is the lidar system constant, $\beta$ is the volume backscatter coefficient (sr km)$^{-1}$, $\sigma$ is the volume extinction coefficient (km$^{-1}$), and $\perp$ and $∥$ represent the planes of polarization orthogonal and parallel to that transmitted. Obviously, the lidar equation in this form has two unknowns, so assumptions are usually made on the relationships between backscattering and attenuation to obtain quantitative data, unless advanced spectroscopic lidar methods (i.e. Raman or high spectral resolution lidar) are used for intrinsic calibration (see Weitkamp 2005). However, to obtain depolarization measurements of the backscatter from linearly polarized transmitted laser light, the linear depolarization ratio is computed simply from the ratio of the backscattered powers in the two orthogonal polarization planes as a function of range (where it is assumed that attenuation affects both signals equally). For two-channel polarization lidars, the ‘total’ linear depolarization ratio is defined by

$$\delta = \frac{P(R)_{∥}}{P(R)_{\perp}} = \frac{[\beta_m(R)_{∥} + \beta_a(R)_{∥} + \beta_c(R)_{∥}] - [\beta_m(R)_{\perp} + \beta_a(R)_{\perp} + \beta_c(R)_{\perp}]}{[\beta_m(R)_{∥} + \beta_a(R)_{∥} + \beta_c(R)_{∥}] - [\beta_m(R)_{\perp} + \beta_a(R)_{\perp} + \beta_c(R)_{\perp}]}$$

(2)

where the subscripts $m = $ molecular, $a = $ aerosol and $c = $ cloud represent the contributions to backscattering from each species. Although the relatively strong backscattering from clouds tends to dominate the total signal, a mixture of molecules and aerosols can produce intermediate $\delta$ values between the value of the pure molecular atmosphere ($\delta \approx 0.03$) and that value characteristic of the aerosol type. Moreover, the size of the aerosol particles relative to the incident laser wavelength also has an impact on the amount of depolarization measured, with the highest depolarization measured for particles equal to or larger than the incident wavelength and lesser amounts generated by smaller particles tending to the Rayleigh scattering regime (Mishchenko and Sassen 1998).

As far as atmospheric smoke studies are concerned, polarization lidar measurements usually reveal near-zero $\delta$ values in fresh smoke layers, as shown in the AFARS lidar height versus time returned laser power (top, based on a logarithmic grayscale) and linear depolarization ratio $\delta$ (bottom, note color scale at right) displays obtained on 13 August 2004. Smoke aerosol is depicted below 4.5 km MSL and in thin layers up to 8.5 km MSL, with scattered cirrus clouds aloft.

**Figure 1.** AFARS lidar height versus time returned laser power (top, based on a logarithmic grayscale) and linear depolarization ratio $\delta$ (bottom, note color scale at right) displays obtained on 13 August 2004. Smoke aerosol is depicted below 4.5 km MSL and in thin layers up to 8.5 km MSL, with scattered cirrus clouds aloft.
depolarization, although ash fallout and droplet crystallization in dry air can lead to higher depolarizations.

3. AFARS research

At AFARS (64°N, −147°W), polarization measurements are possible at 0.532, 0.694, 1.06 and 1.574 μm lidar wavelengths (as well as 3.2 mm wavelength W-band radar), but in this study we rely on findings obtained by the ‘turnkey’ cloud polarization lidar (CPL). This system is based on a high-power (1.5 J), 0.1 Hz pulse repetition rate, ruby laser transmitter and dual backscatter detector channels with 7.5 m range resolution. CPL linear depolarization ratios are thought to be accurate to ±0.02. A photograph of the lidar beneath a roof hatch atop the Geophysical Institute of the University of Alaska Fairbanks is provided in figure 2.

CPL data collection at AFARS began in February 2004 and continues on a regular basis, primarily in support of afternoon satellite overpasses. The targets studied emphasize those most amenable to lidar research, i.e. cirrus clouds, but aerosols and middle-level clouds are also often probed. A graphical and digital data record of the AFARS dataset is compiled at the website, http://rainbow.gi.alaska.edu.

4. Model description

The cloud model used here to simulate the lidar measurements includes explicit spectral bin microphysics for both liquid and crystalline phases with 30 size classes for each phase, along with the supersaturation equation (Khvorostyanov and Sassen 1998a, 2002, Sassen and Khvorostyanov 2007). The model contains six basic units: (1) dynamics (hydrostatic or non-hydrostatic); (2) cloud microphysics (kinetic equations for the droplet and crystal size spectra); (3) thermodynamics (temperature, humidity and supersaturation); (4) aerosols, including transport, deliquescence of the aerosol and nucleation of the liquid and ice phases; (5) longwave and shortwave radiation; and (6) heat and moisture exchange with the underlying surface.

The model accounts for the nucleation of cloud droplets and ice crystals, their transport by wind and turbulence, growth and evaporation by condensation, deposition, coagulation/aggregation and particle sedimentation. The key features for the interpretation of these lidar experiments are the droplet and ice crystal nucleation processes. Parameterization of droplet nucleation was recently upgraded specifically for simulations of aerosol direct and indirect effects. To do this, the Köhler theory was generalized for arbitrary content of the soluble fraction in CCN, and new analytical solutions were found for aerosol hygroscopic growth and drop activation (Khvorostyanov and Curry 2007). The algebraic equivalent was found for the dry and wet lognormal aerosol size spectra, and a generalized power law for the concentration of activated drops \(N_d\) as a function of the maximum water supersaturation \(s_m\) reached in the air parcel, \(N_d(s_m) = C(s_m)k(s_m)^{k(s_m)}\) was formulated which is similar to the Twomey (1959) power law, but both the coefficient \(C(s_m)\) and index \(k(s_m)\) are not constant as in most models but are decreasing functions of \(s_m\) in agreement with experimental data (see Pruppacher and Klett 1997, hereafter referred to as PK97) and are expressed analytically in terms of vertical velocities and CCN microphysical parameters. This allows finite droplet concentrations \(N_d(s_m)\) limited by the total CCN concentration at large \(s_m\), in contrast to many previous theories with unlimited \(N_d(s_m)\).

Although the model includes several schemes of ice nucleation (Khvorostyanov and Sassen 2002), relied on here at these moderately supercooled temperatures is an entirely heterogeneous (i.e. occurring on IN) approach. Parameterization of heterogeneous ice nucleation at such temperatures in this model is based on the theory developed in Khvorostyanov and Curry (2000, 2004a, 2004b, hereafter referred to collectively as KC). In classical heterogeneous nucleation theory, the critical radius \(r_{cr}(T)\) and energy \(\Delta F_{cr}(T)\) of an ice germ, and the nucleation rate \(J_{nuc}(T)\), are functions of temperature only, and any explicit dependence on the relative humidity is absent (PK97). In the KC theory, the classical expressions for the above three quantities are generalized with account taken of the presence of insoluble substrates inside haze particles, and expressed as functions of temperature \(T\) and water supersaturation ratio \(S_w\) simultaneously; then \(r_{cr}(T, S_w)\) and

\[
J_{nuc}(T) = \frac{2\sigma_{is}}{\rho_i L_e^{ef}(T) \ln \left( \frac{1}{4} S_w^2 \right) - C_s, \varepsilon^2 - \frac{T_S}{R} },
\]

with account taken of the ice–substrate misfit strain \(\varepsilon\) and finite size of the germ \(r_d\), and where \(\sigma_{is}\) is the surface tension at the solution–ice interface, \(C_s\) is the Turnbull–Vonnegut constant, \(r_{cr}\) is the scaling radius defined in KC, \(\rho_i\) is the ice density, \(L_e^{ef}(T)\) is the effective melting heat defined in Khvorostyanov and Sassen (1998b), \(T_S = 273.15\) K is the triple point temperature and \(G(T) = RT/(M_e L_e^{mf})\), with \(R\)
being the universal gas constant and $M_w$ the molecular weight of water. The critical energy $\Delta F_{\text{cr,het}} = (4/3) \pi a_n r_n^2 \phi(m_{si})$ includes the Fletcher shape factor $\phi(m_{si})$ that depends on the contact angle or wettability parameter $m_{si}$ of the insoluble substrate and the nucleation rate is expressed as $J_{\text{het}} \sim \exp(-\Delta F_{\text{cr,het}}/kT)$, with $k$ being the Boltzmann constant (PK97). Because this substrate has a catalyzing action, it significantly reduces $\Delta F_{\text{cr,het}}$ and increases $J_{\text{het}}$ compared with the homogeneous case, favoring ice nucleation and shifting the ice nucleation temperature threshold to higher temperatures, usually to $T > -20 \, ^\circ \text{C}$. The invariance relative to chemical composition is absent in equation (3) because the critical energy and nucleation rate depend on $m_{si}$, which determines the shape factor of an ice germ. All these effects depend on the chemical composition of the substrate. Note that it was shown in Khvorostyanov and Curry (2004b) that this approach justifies the methods based on the concept of the effective freezing temperature introduced in Sassen and Dodd (1988) for homogeneous nucleation, as since used in many works and applied in DeMott et al. (1997) for heterogeneous nucleation. Thus, ice formation in cirrus and supercooled liquid clouds could be closely related.

Although the chemical nature of mixed CCN arising from forest fire plumes is not well understood, the parameter $m_{si}$ given in table 5.2 in PK97 for surface soil ($m_{si} = 0.36$–0.42) and quartz ($m_{si} = 0.63$–0.72) might be appropriate, while for soot Kärcher et al. (1996) measured $m_{si} = 0.44$–0.57. Both crustal materials elevated in fire convective plumes, and carbonaceous soot formed by burning, may constitute the insoluble fraction of smoke particles that serve as IN. Therefore in this study average values of $m_{si} = 0.48$ or 0.50 were taken to simulate the heterogeneous ice nucleation of growing aqueous smoke droplets becoming diluted in updrafts. It is also hoped that this approach might serve to mimic the nucleation of ice in growing aqueous solutions of certain organic substances without solid IN, if indeed this is taking place, though the framework to treat this process is currently unavailable (Cantrell and Heymsfield 2005).

The model can be configured in 1D, 2D or 3D versions. For this study, we use the 1D model version similar to Sassen and Khvorostyanov (2007) in the following manner. The main computational domain is located at heights from 3.0 to 6.0 km in the vertical and included 61 vertical grid points with 50 m resolution. For radiative calculations, an additional seven levels are used below the main domain (0–3.0 km) with resolution $\Delta z = 0.5 \, \text{km}$ and nine levels above (6.0–24.0 km) with $\Delta z = 2.0 \, \text{km}$, which is sufficient in the cloudless atmosphere. The model was initialized with sounding data corresponding to one of the lidar case studies, as described below. The boundary conditions were chosen in the form of zero vertical turbulent fluxes at the lower and upper boundaries of the main domain. The size spectra of the deliquescent aerosol occurring due to fires are still highly uncertain, so in this study they were chosen in algebraic form, equivalent to lognormal with a mean geometric radius of 0.02 \( \mu \text{m} \) and dispersion 2.5. The concentration of the cloud condensation nuclei was $N_{\text{CCN}} = 150 \, \text{cm}^{-3}$, which was constant in the middle domain, and that of the ice nuclei was $N_{\text{IN}} = 5.0 \, \text{cm}^{-3}$, which is meant to imitate an increase in ice nuclei due to smoke.

5. AFARS case studies

In this section we provide two case studies of apparent smoke/liquid cloud interactions collected during the severe 2005 summer fire season in the interior of Alaska. These examples show supercooled altocumulus clouds forming at the top of ~5.0 km deep smoke-filled boundary layers during times when a combination of fresh local and more aged smoke aerosol was present. (During some periods Fairbanks was enclosed in smoke so dense that lidar operations were impracticable due to severe optical attenuation and the risk of laser component damage from aerosol deposits.) As in figure 1, included in the lidar height–time displays are the local sounding data.

Figure 3 presents CPL data from the afternoon of 3 August 2005 as cirrus clouds from 7.0–10.5 km MSL were moving through central Alaska and mid-level altocumulus were developing locally at 5.0 km MSL in association with afternoon cumulonimbus activity. The numerous cloud droplets in the altocumulus layer, when present, produce strong laser backscattering and attenuation, which blocks the returns from the cirrus clouds aloft. The MODIS satellite image obtained at 2210 UTC in figure 4 reveals several plumes of smoke from fires surrounding Fairbanks (the arrow), the cirrus approaching from the south and the cellular altocumulus clouds. At top right can be seen an anvil developing from cumulus convection, where the altocumulus appear to be spreading radially in response to gravity waves.
Figure 4. The Aqua satellite MODIS true color image collected at 2210 UTC on 3 August 2005 corresponding to the lidar data in figure 3. Shown are invading cirrus clouds (bottom right), developing cellular altocumulus clouds and smoke plumes surrounding the Fairbanks AFARS site (see arrow).

Figure 5 presents an expanded view of the lidar images below 6.0 km MSL. These show more clearly that the supercooled (∼−15°C) liquid layer is first detected at 2312 UTC and generates near-zero \( \delta \) at cloud base but with increasingly higher \( \delta \) (up to ∼0.20) as the laser pulse penetrates into the dense cloud due to the action of photon multiple scattering activity. However, it is clear that the returns from ice particles begin below the 5.0 km MSL liquid cloud base height about 10 min before the liquid layer is observed. In this case, the ice returns are not significantly depolarized like in most of the cirrus clouds aloft because the ice plate crystals were mostly horizontally oriented, producing specular reflections and thus no depolarization when viewed by the lidar in the zenith direction (e.g. Sassen 2005). (Operationally, this effect vanishes when the lidar is tilted a few degrees off the zenith direction, as is routinely tested in the field.) Nonetheless, it is important to emphasize that this appearance of ice virga before the parent liquid cloud is highly unusual in lidar data. The normal situation is that the liquid cloud layer appears first and then generates ice virga that gradually descends to lower levels with time (e.g. Sassen and Khvorostyanov 2007). It can also be seen that intermittent, thin ice plate layers also occur earlier at about the same heights without the presence of supercooled droplet clouds. This too suggests the nucleation of ice below water saturation.

Figure 6 illustrates another AFARS example from a similarly supercooled altocumulus cloud in the presence of smoke aerosol studied on 11 June 2005. Cirrus clouds are present initially from ∼8.0 to 9.0 km MSL, and also as a subvisual layer from 10.0–11.5 km MSL just below the tropopause. As shown more clearly in the expanded lidar displays in figure 7, oriented ice plates are again present around 4.5 km MSL starting 10–15 min before the liquid layer first becomes evident at 2222 UTC. In this case, however, the liquid layer periodically glaciates, leaving only ice crystals behind. These periods correspond to those times when the lidar pulse is able to reach the cirrus cloud in figure 6, because of the decreased optical attenuation encountered as a result of the phase change, and can also be recognized because of the lack of cloud droplet multiple-scattering-induced depolarization in figure 7. When the altocumulus layer is present, an
intense, narrow laser return is visible at $\sim 5.0$ km MSL and the depolarization data below are restricted because the lidar detector gains were reduced to enable the capture of the strong liquid cloud signals. The smoke in this case appears to have been generated by major forest fires in southeastern Alaska.

### 6. Model simulation results

The model was initialized using the sounding profiles from Fairbanks, Alaska, on 4 August 2005 at 0000 UTC. Figure 8 shows the profiles of temperature (a), relative humidities (b) over water ($\text{RH}_{w}$, solid line) and ice ($\text{RH}_{i}$, dashed line), wind speed (c) and wind direction (d) that correspond to the lidar display in figure 3. The temperature profile exhibits a layer with increased stability just above a height of 5.0 km. This stable layer is similar to the inversion-capped boundary layer and is situated in a region of strong directional wind shear and a local maximum in wind speed. It is also associated with a near-water-saturated $\text{RH}_{w}$ peak, implying moisture advection that would accelerate the dilution of smoke-derived haze particles. According to equation (3), this combination of increased $\text{RH}_{w}$ ($\geq 90\%$) with sufficiently cold temperature ($T \approx -15^\circ C$) is favorable for heterogeneous ice nucleation in the immersion or deliquescent-freezing mode.

Since we use a 1D single-column model, where explicit advection is absent, the method usually applied in single-columns models is used: i.e. the prescription of some value of advection (e.g. Randall and Cripe (1999), Jiang et al (2000), Sassen and Khvorostyanov (2007)). It was found in several numerical experiments that a parabolic profile of humidity advection ($dq/dt_{\text{adv}}$ in the relatively thin layer 4.4–5.2 km (that imitates the advection rate seen in figure 8) was
Figure 9. 1D cloud microphysical model results of mixed-phase cloud formation and development for the baseline (peak 
\( dq/dt \) adv, m \( \approx 0.18 \) g kg\(^{-1}\) h\(^{-1}\) and \( m_{sw} = 0.48 \)) 3 h model run. Shown are height versus time displays of water and ice supersaturation ratios, crystal and droplet concentrations, ice water (IWC) and liquid water (LWC) contents, and crystal and droplet mean radii.

suitable in this case. A maximum value of \( (dq/dt)_{adv,m} \approx 0.18 \) g kg\(^{-1}\) h\(^{-1}\) was chosen. This value is comparable to those used in earlier works and ensures the formation of an altocumulus cloud in less than 2 h. This run is called below the ‘baseline’, whereas the values of \( (dq/dt)_{adv} \) were varied in other model runs. In addition, a slight cooling rate was imposed by introducing a parabolic profile of updrafts in the same layer with a maximum of 1.5 cm s\(^{-1}\) at the height of maximum humidity advection.

Figure 9 shows height–time displays of the cloud formation and development with imposed \( (dq/dt)_{adv} \) leading to the humidification of the layer. The critical saturation ratio for heterogeneous freezing at the \(-15^\circ\)C temperature is reached first and causes the formation of the elevated layer of the crystals in clear sky. Crystals begin to form at about 95 min at slight subsaturation, \( S_{sw} \approx 0.97 \) (RH\(_{sw} = 97\%\)) at a height of 5.0 km due to the heterogeneous freezing of deliquescent smoke-haze particles with contact parameter \( m_{sw} = 0.48 \).

The key to understanding the physical mechanism of their formation is equation (3), which shows how both \( T \) and \( S_{sw} \) influence this process.

Subsequently, the rate of supersaturation generation due to advective humidification and convective cooling by updrafts still exceeds the rate of vapor absorption by the crystals, and relative humidity continues to grow. Water saturation is reached and droplets first nucleate at 110 min, i.e. the liquid phase begins to form \( \approx 15 \) min after the first crystals nucleate, similar to the lidar observations in figures 3 and 6. The ice
Crystal generation layer is located during the run in a narrow layer 5.0–5.1 km, the crystals grow in and precipitate from the ice supersaturated layer to 4.5 km, and then gradually evaporate beneath. The liquid layer remains near 5.0 km. The superposition of the ice and liquid water fields is shown in figure 10 in a form similar to that revealed in the lidar images (compare to figure 5), and so illustrates the time lag of liquid phase formation relative to the ice phase.

Vertical profiles of microphysical properties after 3 h of simulation are given in figure 11. Panels (a) and (b) illustrate the initial profiles of supersaturations with respect to water $\delta_w$ and ice $\delta_i$. (This justifies our choice of humidity advection rate since after 3 h the profiles are similar to the initial ones, but $\delta_i$ is close to zero near 5.0 km in the liquid layer while $\delta_i$ reaches a maximum 20%.) Crystal concentrations reach $1.3 \times 10^5$ in the upper layer near 5.0 km and decrease downward in the ice subsaturated layer; the maximum ice water content (IWC) is 2.0 mg m$^{-2}$ at 4.5 km where $\delta_i = 0$; mean crystal radii are 20–40 $\mu$m at 5.0 km and grow downward until reaching 80–100 $\mu$m at the bottom of the crystalline layer. Thus, the crystal layer evaporates very slowly over the $\sim 2.0$ km distance. In contrast, the liquid layer is thin and concentrated near the maximum of the humidity advection rate; maxima of droplet concentration and liquid water content (LWC) are 80 cm$^{-3}$ and 0.09 g m$^{-3}$, respectively, with droplet radii of 4.0–7.0 $\mu$m.

Because it was found in the parcel model study of Khvorostyanov and Curry (2005) that this heterogeneous ice nucleation scheme is sensitive to the chosen value of $m_{iw}$, figure 12 shows similar height–time displays using a contact parameter $m_{iw} = 0.50$. The general picture of cloud formation is similar to the previous run, but the maximum value of crystal concentrations is higher and reaches 8.4 l$^{-1}$. The value of IWC is correspondingly higher, and LWC, lower. There are still substantial uncertainties in the measured values of the contact parameter (PK97) and this comparison indicates the importance of such measurements.

Numerical experiments varying the other parameters have shown that ice nucleation is sensitive to the humidity advection rate, which may account for the lidar observation of ice crystals being detected without a supercooled parent cloud. Figure 13 shows simulation results for the case with an increased width of humidity advection layer (4.2–5.6 km versus 4.4–5.2 km in the baseline run) and using a maximum vertical velocity $w_m = 1$ cm s$^{-1}$ and contact parameter $m_{iw} = 0.5$. The crystalline layer forms at 70 min (25 min earlier than in the control run) and the liquid phase, paradoxically, does not form at all, because: (a) increased humidity advection at higher altitudes causes faster humidity growth above 5.0 km (i.e. at colder temperatures) and (b) the critical saturation ratio is lower at colder temperatures, which allows earlier nucleation. The crystal concentration reaches 10–15 l$^{-1}$ in the narrow layer at 5.0–5.2 km, where ice nucleation at $S_w = 0.97–0.99$ takes place from 70 min to the end of the run. The supersaturation budget is such that its generation is balanced by crystal absorption, so that the maximum value reaches $-0.5$ to $-1\%$ and is equilibrated at this level. Thus, water supersaturation is not attained and a liquid phase cloud is not formed. Such elevated layers caused by aerosols and humidity advection may persist for a few hours or days (e.g. Curry et al 1996).

7. Discussion and conclusions

Polarization lidar is a tool that has demonstrated capabilities to research aerosol/cloud interactions, in that the technique can detect and often distinguish between many cloud particle and aerosol types. Even simple two-channel lidar systems are quite useful in this regard, although more sophisticated multi-channel systems can provide more quantitative data on a range of scattering/attenuation properties (see Weitkamp 2005). In recent years it has been shown that transported desert dusts serve in the atmosphere as effective IN, thereby affecting cirrus cloud formation (Murayama et al 2001, Sassen 2002) and supercooled cloud phase (Sassen et al 2003). In this study we have examined, for the first time, polarization lidar data indicating the effects of forest fire smoke on supercooled water droplet cloud formation. Clearly, smoke-induced changes in cloud contents and phase may affect the radiation balance of the earth/atmosphere system, and as biomass smoke increases due to human activities, the implications of this potential climate feedback mechanism need to be evaluated.

Interestingly, it has long been recognized that certain organic substances are efficacious IN in the laboratory (Fukuta 1966), while field data also show that forest fire smoke (Hobbs and Locatelli 1969) and sugar cane fires (Pueschel and Langer 1973) are capable of generating IN. Only recently have laboratory studies begun to revisit the question of the ice nucleating capabilities of the products of biomass burning, including soot aerosols coated by various materials (DeMott 2002, Möhler et al 2005), and aqueous long chain alcohol
solutions (Cantrell and Robinson 2006). Another possibility is that soil particles raised by fires and then embedded in droplets can serve as IN, because smoke aerosol can contain considerable mineral content (PK97). In all three scenarios, immersion ice nucleation activity can be expected to be a function of relative humidity, which controls the solution strength of the growing haze droplets, the precursors to cloud droplets in updrafts. A similar mechanism is believed to prevail in cirrus clouds, where ice crystals form homogeneously in growing haze particles with diluted solution strengths. Deposition nucleation, which involves ice growth on dry particles directly from the vapor, could also mimic the

Figure 11. Vertical profiles of mixed-phase cloud microphysical properties from figure 9, but for results obtained at 3 h of simulation time.
Figure 12. Height–time mixed-phase cloud model prediction similar to figure 9 but using a slightly increased water vapor advection rate and $m_{is} = 0.50$ inputs.

production of the ice crystals observed by the lidar below water saturation, but in view of the polluted environmental chemistry accompanying smoke aerosol formation, this mechanism is unlikely to be significant.

The polarization lidar measurements exemplified here have revealed an odd sequence of events connected to supercooled altocumulus clouds in contact with boreal smoke aerosol. Ice crystals that appear before the water cloud, or at the same sub-cloud heights without evidence for the liquid phase, have been noted in several cases. In contrast to the usual sequence of ice virga following the initial appearance of the liquid layer, the ice crystal virga appears first. So, the ice crystals are not virga in the sense that they sedimented from the liquid layer after nucleation therein, but rather they formed in updrafts in water subsaturated air. These ice crystals are often in the form of horizontally oriented plates (as in figures 4 and 7), but this should not be surprising at temperatures of $\sim -15^\circ C$ in the middle of the planar ice crystal growth regime. One example is given that indicates the intermittent glaciation of a supercooled liquid cloud, whereas this amount of supercooling can be considered as moderate. Note, for example, that the mean temperature of mid-latitude altocumulus from lidar measurements was about $-15^\circ C$, which could produce ice virga, but some altocumulus clouds have been observed to approach the $-40^\circ C$ homogeneous freezing temperature before glaciation occurs (Sassen and Khvorostyanov 2007).

Insights from the model simulations tailored to the field data support the concept that sub-cloud (i.e., sub-water-saturated) ice nucleation can result from the activation of IN in smoke droplets undergoing the Köhler swelling process in updrafts. The lidar data also indicate that water cloud formation may or may not occur and that portions of the liquid layer when present may glaciate. Varying the model conditions indicates that these various indirect cloud effects can result from the specifics of the IN composition, especially the $m_{is}$ value, or the atmospheric dynamics like the humidity advection rate. The most unusual feature, apparently characterized here for the first time, is the reverse order of formation of the ice and liquid phases associated with mixed-phase clouds: the ice forms earlier than the liquid phase by the delayed immersion freezing process below the liquid cloud base. Thus the model has been able to explain the lidar findings, given certain boundary conditions, based on the generalization of classical nucleation theory.

Although the formation of ice 10–20 min earlier than the liquid cloud was also found in the parcel model runs of Khvorostyanov and Curry (2005) based on the heterogeneous nucleation scenario, the lidar measurements in the current case dramatically illustrate that such effects do occur in the atmosphere at temperatures of $\sim -15^\circ C$ in the presence of smoke aerosol. An explanation of this process cannot be made in terms of the classical heterogeneous ice nucleation theory (PK97), which contains the temperature factor but not the humidity factor. Rather, the experimental results can be simulated using the generalizations of the classical theory that are similar for homogeneous (Khvorostyanov and Sassen 1998b) and heterogeneous ice nucleation (KC). Interestingly, the similarity of these theories in terms of critical ice germs shows that ice formation associated with cirrus and supercooled altocumulus clouds can involve related processes.

To better understand the possible effects of biomass burning, what is needed is a wide range of laboratory and field studies of smoke content and its ice nucleating capabilities. Not only is the composition of smoke aerosol likely to differ considerably with the local biomass type and soil characteristics, but even in boreal smoke there are likely to be a variety of possible cloud effects. Fires surrounding the
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Figure 13. Height–time displays of ice cloud formation and development (without the liquid phase) using an increased depth of humidity advection (4.4–5.6 km versus 4.4–5.2 km in the baseline run), maximum vertical velocity of 1.0 cm s$^{-1}$ and contact parameter $m_{\text{cr}} = 0.50$.

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