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Three examples where the specific surface area of snow increased over time

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Abstract. Snow on the ground impacts climate through its high albedo and affects atmospheric composition through its ability to adsorb chemical compounds. The quantification of these effects requires the knowledge of the specific surface area (SSA) of snow and its rate of change. All relevant studies indicate that snow SSA decreases over time. Here, we report for the first time three cases where the SSA of snow increased over time. These are (1) the transformation of a melt-freeze crust into depth hoar, producing an increase in SSA from 3.4 to 8.8 $m^2\,kg^{-1}$. (2) The mobilization of surface snow by wind, which reduced the size of snow crystals by sublimation and fragmented them. This formed a surface snow layer with a SSA of 61 $m^2\,kg^{-1}$ from layers whose SSAs were originally 42 and 50 $m^2\,kg^{-1}$. (3) The sieving of blowing snow by a snow layer, which allowed the smallest crystals to penetrate into open spaces in the snow, leading to an SSA increase from 32 to 61 $m^2\,kg^{-1}$. We discuss that other mechanisms for SSA increase are possible. Overall, SSA increases are probably not rare. They lead to enhanced uptake of chemical compounds and to increases in snow albedo, and their inclusion in relevant chemical and climate models deserves consideration.

1 Introduction

The snow cover is an interface that probably has one of the highest impacts on the exchanges of energy and chemical species between the atmosphere and the surface of the Earth (Domine et al., 2008). One crucial snow physical variable relevant to these processes is its specific surface area (SSA), defined as the surface area accessible to gases per unit mass (Legagneux et al., 2002). The SSA is closely related to the surface (S) to volume (V) ratio of snow crystals used in models that describe the interactions between snow and optical radiation (Warren, 1982; Kokhanovsky and Zege, 2004). If snow crystals are assumed to be spheres, then SSA=$S/\rho V$=$3/(\rho r)$, where ρ is the volumic mass of ice and r the radius of the spheres representing the snow crystals.

Because snow has a high SSA, up to 156 $m^2\,kg^{-1}$ for fresh snow (Domine et al., 2007a), snow scatters light, is highly reflective and its albedo increases with increasing snow SSA (Warren, 1982; Domine et al., 2006). The high albedo of snow helps cool the Earth's surface. In current climate warming scenarios snow cover is expected to decrease (Dye, 2002; Stone et al., 2002; Pielke et al., 2004). The replacement of snow by darker surfaces (exposed vegetation, soils or ground cover) explains why polar regions are most affected by warming (Hall, 2004).

Snow is also known to adsorb chemical species such as semi-volatile organic compounds (SVOCs) in amounts proportional to snow SSA (Wania et al., 1998; Daly and Wania, 2004; Herbert et al., 2005; Domine et al., 2007b), and this affects atmospheric composition. Changes in snow SSA caused by snow metamorphism (Taillandier et al., 2007) result in exchanges of SVOCs between the atmosphere and the snow. In particular, upon snowmelt, chemical species can be released to the atmosphere, resulting in a sudden surge in their atmospheric concentrations (Daly and Wania, 2004). Species stored by the snowpack can also be released to terrestrial and marine ecosystems (Meyer et al., 2006).

Modeling these climatic and chemical processes requires the parameterization of snow SSA as a function of time. All available experimental and modeling studies have shown that snow SSA decreases with time (Cabanes et al., 2002, 2003; Legagneux et al., 2003, 2004; Legagneux and Domine, 2005;

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Flanner and Zender, 2006; Taillandier et al., 2007). This is expected, since a high SSA produces a high surface energy per unit mass, which is thermodynamically unstable. This SSA decrease predicts that snow albedo and the storage capacity of adsorbed chemical species will decrease with time.

However, snow processes are not ruled by thermodynamics only and snow SSA can in principle increase, for example if energy is transferred to the snow or if water vapor is added or removed by condensation or sublimation. Even though instances of such processes resulting in SSA increases have not been reported, we have observed them on a number of occasions and detail three examples here. Their inclusion in models where snow is a component may help explain or predict processes such as the enhanced uptake of chemical species by snow or increases in snow albedo in the absence of precipitation (Liljequist, 1956; McGuffie and Henderson-Sellers, 1985; Pirazzini, 2004).

The three examples where the SSA of snow increased that we report here are: (1) the transformation of a melt-freeze crust into depth hoar; (2) the mobilization of surface snow by wind, which reduced the size of snow crystals by sublimation and fragmentation; (3) the sieving of blowing snow by an unperturbed snow layer, leading to the deposition of the smallest crystals inside the layer.

2 Methods

2.1 Snow sampling

The sampling procedure employed in this study has been described earlier (Hanot and Domine, 1999 and Domine et al., 2002). Briefly, a new snow pit with vertical faces was excavated for each sampling event to observe the stratigraphy and identify the layers of interest. Density was measured by weighing a horizontal core of known volume. This method has an accuracy of about 5% for layers thicker than 3 cm. For SSA measurements, about 100 cm^3 of snow was collected in a glass vial with a stainless steel spatula. The vial was immediately immersed in liquid nitrogen to stop metamorphism until its content was transferred to the SSA measurement container in a cold room at a temperature below $-15°C$. When possible, photomicrographs of the snow samples were taken under reflected light with a reflex camera fitted with bellows and a 35 mm macro lens.

2.2 Measurements of the specific surface area of snow

Snow SSA was determined by measuring the adsorption isotherm of methane at liquid nitrogen temperature (77 K), as detailed in Legagneux et al. (2002). A mathematical treatment was applied to the adsorption isotherm to derive the SSA. The method has a reproducibility of 6% and an accuracy better than 12% (Legagneux et al., 2002). Since the work of Legagneux et al. (2002) an experimental artifact due to CH_4 adsorption on the stainless steel walls of the snow container was detected, as detailed in Domine et al. (2007a). All data presented here are corrected for this artifact.

3 Results

3.1 Transformation of a melt-freeze crust into depth hoar

The first case study occurred during the winter of 2003/2004 at the Large Animal Research Station (LARS) of the University of Alaska Fairbanks (64°52′ N, 147°44′ W). The detailed study of the evolution of the snowpack has been presented by Taillandier et al. (2006). However they did not report the increase in SSA that we present here based on their complete data. The onset of snowpack formation took place on 25 October. The focus of this work is the snowfall of 7 November. For 2 days, it was subjected to temperatures that rose above 0°C (Fig. 1a), and to very light rain, resulting in the formation of an 8 mm thick melt-freeze crust on 9 November, which was fairly solid and looked completely impermeable to the eye. Temperatures then rapidly dropped below freezing, to reach $-37°C$ on 17 November. The SSA of that melt-freeze crust was measured on three occasions (Fig. 2), and the snow crystals were photographed each time (Fig. 3).

Initially, the melt-freeze layer consisted of large well-connected grains. Its SSA was $3.4\,m^2\,kg^{-1}$ on 11 November. In interior Alaska, thin snowpacks and cold temperatures combine to produce very large temperature gradients (Fig. 1b), that generate large upward water vapor fluxes. These favor the growth of large depth hoar crystals (Sturm and Benson, 1997; Taillandier et al., 2006) which in this case were sufficient to transform this hard impermeable layer into depth hoar. Figure 3 shows that on 27 January the layer was comprised of a mixture of depth hoar and melt-freeze crystals, with a SSA of $8.5\,m^2\,kg^{-1}$. On 12 February, it was comprised mostly of depth hoar crystals, with a SSA of $8.8\,m^2\,kg^{-1}$. Given the measurement reproducibility of 6%, and the fact that small spatial variations in SSA may exist (Domine et al., 2002), these last 2 values are essentially identical, and consistent with observations that the terminal SSA of depth hoar in the subarctic snowpack around Fairbanks is about $8\,m^2\,kg^{-1}$ (Taillandier et al., 2006). With these transformations, the cohesion and hardness of this layer decreased significantly: a 10 cm long slab of this layer would break under its own weight when held in the air. Until snow melt, however, it remained noticeably harder than the over- and underlying depth hoar layers.

3.2 Remobilization of surface layers by wind

This case study took place in February 2000 near Alert, in the Canadian high Arctic (82°27′ N, 62°30′ W). Two snowfalls took place on 3 and 7 February, as described in Cabanes et al. (2002) and Domine et al. (2002). They consisted of small columns and bullet rosettes (Fig. 4) and had SSAs

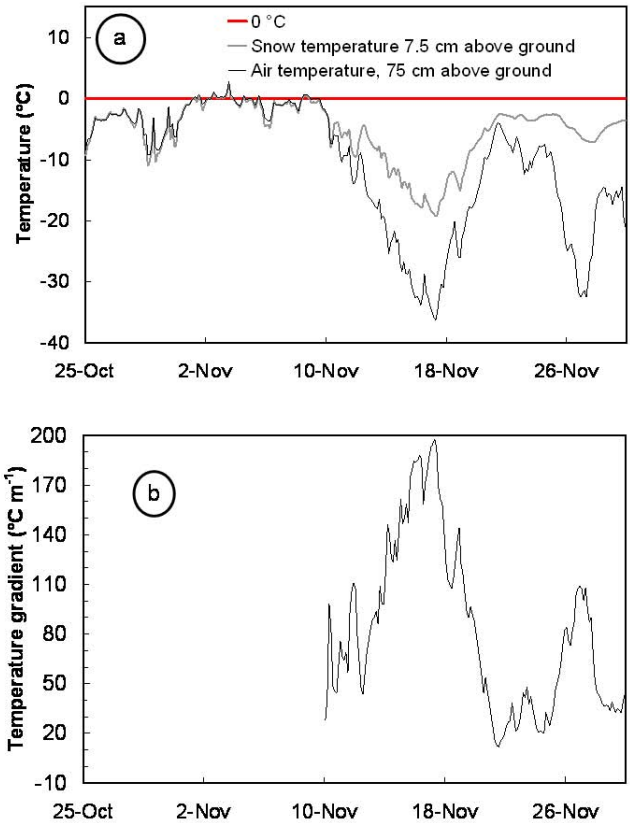

Fig. 1. (a) Temperature of snow and air at our study site near Fairbanks, Alaska, during the 2003–2004 winter. Temperatures at 7.5 and 75 cm above ground were almost the same until the lower sensor was buried by snow on 10 November. **(b)** Temperature gradient in the Alaska snowpack at the same dates, determined from differences in temperature between a sensor on the ground and a sensor 7.5 cm above ground. Data start when the upper sensor was covered by snow on 10 November.

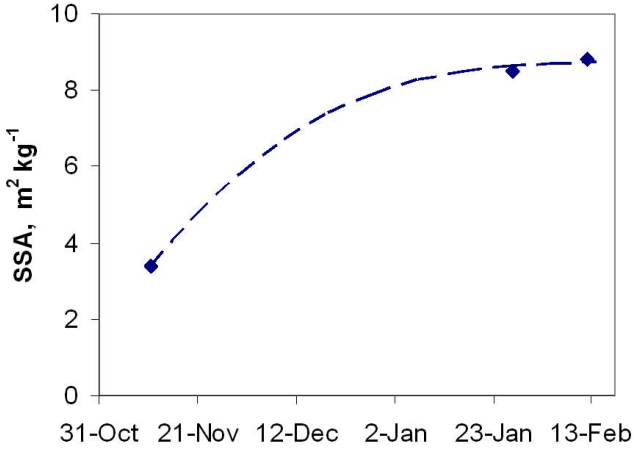

Fig. 2. Temporal evolution of the specific surface area of the melt-freeze crust studied in Alaska. The fit is our estimate, based on our observations that transformation to depth hoar is fastest early in the season, when the snowpack is thin and the temperature gradient highest.

measured one day after they fell of 73.4 and 140.1 $m^2\,kg^{-1}$, respectively. The 7 February layer completely covered the 3 February layer and was itself subjected to the deposition of surface hoar. The SSA of both layers decreased over time. The SSA decrease of the 3 February layer is explained by crystal rounding and the disappearance of small structures under low temperature gradient conditions, while that of the 7 February layer is explained by both crystal rounding and dilution by surface hoar of lower SSA (Fig. 4). The SSA of surface hoar could not be measured, as these crystals formed too thin a layer to be collected separately. The SSA of hoar frost that was growing on antenna guy wires could nevertheless be measured, 54 $m^2\,kg^{-1}$, and those crystals looked similar to those of the surface hoar.

Figure 5 shows that the SSA of both layers decreased monotonically, as expected, until 20 February. On 21 February, a wind storm started in the absence of precipitation. Both snow layers were remobilized, mixed and drifted because of wind action. They accumulated in spots sheltered from the wind, forming a discontinuous layer with sastrugi, over an older windpack that was now partly exposed. That newly formed discontinuous layer was sampled on 22 February and had a SSA of 61 $m^2\,kg^{-1}$ (Fig. 5), higher than those of previous layers 2 days before (42 and 50 $m^2\,kg^{-1}$). The new layer consisted of small rounded grains, produced by the sublimation of the small grains derived from the precipitated bullet rosettes and columns, and of rounded surface hoar crystals (Fig. 4). The SSA increase is presumably due to sublimation of grains while airborne, which reduced their size and hence their SSA, and to fragmentation, which created new surfaces.

Metamorphism, in particular induced by temperature gradients, could also produce SSA changes. However, such processes do not significantly affect SSA on timescales of a day (Taillandier et al., 2007) except for fresh snow of very high SSA. Furthermore, metamorphism would have led to a decrease in snow SSA (Taillandier et al., 2007; Cabanes et al., 2003), not to an increase as observed here.

3.3 Sieving of blowing snow by a snow layer

The last case study took place in February 1999 at Col de Porte (45°12′ N, 5°44′ E), at an altitude of 1320 m, just north of Grenoble in the French Alps. Sampling took place within the study site of the Centre d'Etude de la Neige (Météo-France). The snow layer of interest fell between 6 and 9 February, and had a thickness of 105 cm on 10 February. Some aspects of this snowfall are reported in Cabanes et al. (2003) but the event that resulted in the SSA increase was not mentioned.

Fig. 3. Photomicrographs illustrating the formation of depth hoar from the melt-freeze crust studied in Alaska. Samples are from (left to right) 11 November 2003, 27 January 2004 and 12 February 2004. Scale bars: 1 mm.

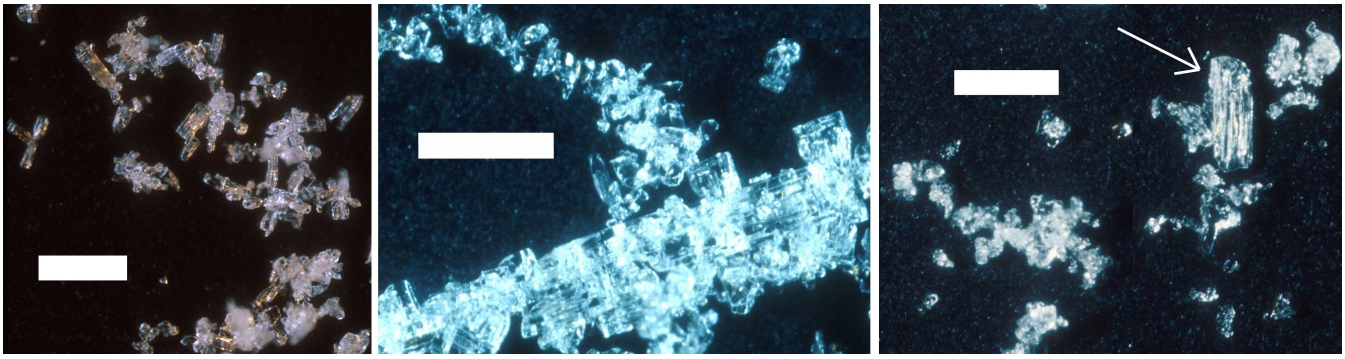

Fig. 4. Photomicrographs showing the evolution of surface snow at Alert in February 2000. From left to right : 3 February layer on 4 February, showing bullet rosettes and columns; 7 February layer on 20 February, showing very rounded precipitated crystals and well developed surface hoar crystals; both layers mixed by wind on 22 February, showing rounded and sublimated crystals. A rounded surface hoar crystal is clearly visible in the top right (arrow). Scale bars: 1 mm.

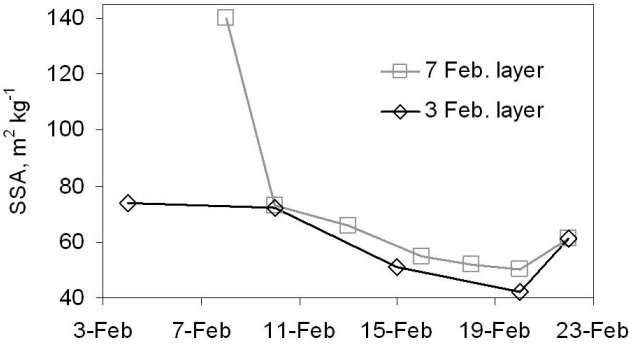

Fig. 5. Evolution of the specific surface area of two surface snow layers near Alert, Canadian Arctic. Until 20 February, the SSA decreased monotonically as expected. The 21 February windstorm remobilized the snow, grains sublimated and fragmented, leading to an SSA increase.

Figure 6 shows the evolution of the stratigraphy of the layer studied. The first sample collection occurred on the morning of 10 February. Around 16:00 LT on that day, the wind picked up and remobilized the snow. The wind lasted until the early morning of 12 February, and eroded the upper half of the snow layer. 25 cm of drifted snow accumulated on top of the remaining half of the layer. Figure 7 shows the SSA evolution of the two sublayers indicated in Fig. 6.

The feature of interest is the increase in SSA of sublayer (a) from 32.3 to 60.5 $m^2\,kg^{-1}$. Unfortunately, no photomicrographs were taken at the time. However we provide the simplest possible explanation of the mechanisms related to this increase. We suggest that the smallest crystals of the drifting snow were trapped into sublayer (a) during the windstorm, and the addition of these small particles with a high SSA produced the observed increase. Wind blowing over the rough surface of a porous solid such as snow produces wind pumping inside the snow (Waddington et al., 1996) and ventilation can extend down to depths of several tens of cm (Sokratov and Sato, 2000). Wind pumping can deposit aerosol particles inside the snow, not just on its surface (Cunningham and Waddington, 1993; Waddington et al., 1993; Harder et al., 2000; Domine et al., 2004). We suggest that the snow acted like a sieve: the largest drifting crystals remained on the surface while the smallest ones could penetrate inside the snow just like aerosol particles, down to depths of several cm. The density of sublayer (a) was 180 $kg\,m^{-3}$ both on 10

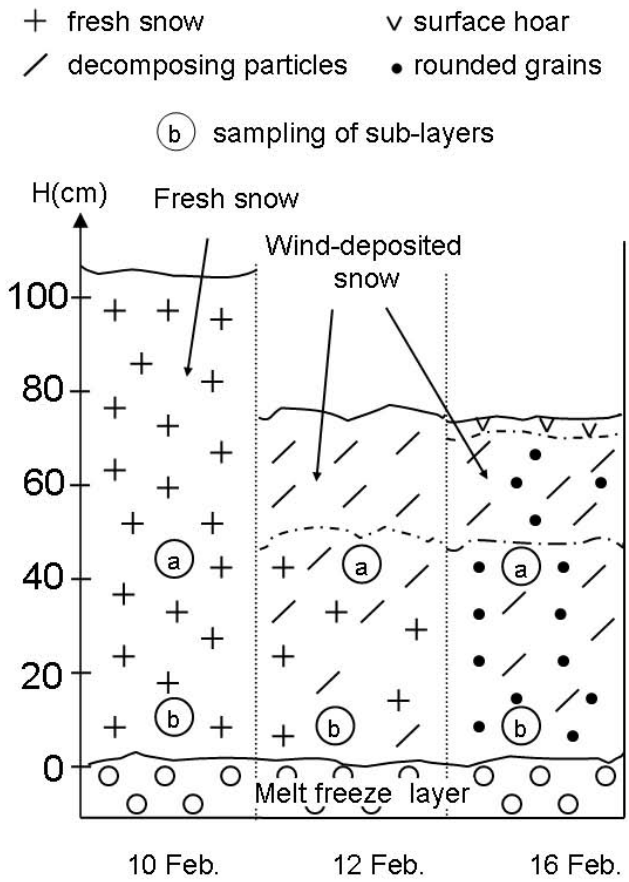

Fig. 6. Col de Porte stratigraphy in February 1999, showing sampling locations.

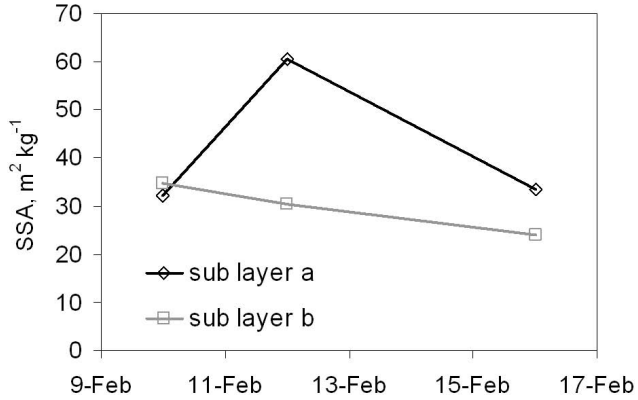

Fig. 7. Evolution of the specific surface area of two sublayers of the surface snow layers at Col de Porte, French Alps. This illustrates the SSA increase in sublayer a between 10 and 12 February 1999, thought to be due to sieving of blowing snow by sublayer a.

and 12 February, meaning that the porosity was >80%. This highly porous snow likely facilitated the transport of small crystals into the snow layer.

Other explanations are possible. There may have been a SSA discontinuity in the stratigraphy, since the snowfall consisted of distinct events. Successive samplings on either side of a discontinuity would produce SSA variations. However, visual examinations with an 8× magnifying glass revealed a fairly homogeneous layer comprised of plates, needles and columns with little or no rime. Further studies of similar cases are nevertheless needed to confirm that the sieving of blowing snow can lead to the penetration of the smallest crystals inside underlying layers.

4 Discussion

The SSA of snow in a closed system and under isothermal conditions decreases with time because of basic thermodynamic considerations that predict that the surface energy will decrease. Under temperature gradient conditions, the SSA decrease of precipitated snow is accelerated because of more vigorous water vapor fluxes (Flanner and Zender, 2006; Taillandier et al., 2007). We show here that when a strong temperature gradient is applied to snow of low SSA, or when snow is remobilized by wind, its SSA can increase, with possible consequences on the fluxes of energy and chemical species. It is therefore of interest to discuss the frequency of such events.

4.1 Frequency and impact of the SSA increase of melt-freeze crusts

The transformation of a melt-freeze crust into depth hoar indicates that any snow type subjected to a sufficiently strong temperature gradient will transform into depth hoar. This is in disagreement with early studies (Akitaya, 1974; Marbouty, 1980) suggesting that depth hoar could not form in dense snow such as melt-freeze crusts. However, Sturm and Johnson (1991) reported that refrozen layers in the subarctic snowpack quickly metamorphose into depth hoar, and Sturm and Liston (2003) reported the transformation of hard wind slabs into depth hoar, confirming that dense layers can be transformed into depth hoar under extreme temperature gradients. In the case studied here, depth hoar above and below the melt-freeze crust, formed directly from precipitated crystals, had essentially the same SSA, around $8\,m^2\,kg^{-1}$ (Taillandier et al., 2006), as the transformed melt-freeze crust, suggesting that the SSA of depth hoar is not affected by the initial nature of the snow. At present, we lack data to quantitatively evaluate the frequency of the transformation of melt -freeze layers into depth hoar or faceted crystals. However, melt-freeze layers are frequent in the Arctic. We have observed them in Northern Alaska and in Svalbard every spring that we were there.

Regarding mid-latitudes, Birkeland (1998) reports that radiative heating can induce the formation of a melt layer a few mm below the snow surface. With this layer acting as a source of vapor, and elevated transient temperature gradients (up to 200°C m^{-1}, Birkeland et al., 1998) caused by diurnal temperature cycles, Birkeland observed the formation of thin layers of faceted crystals just above the melt layer. Even though Birkeland did not study the transformation of the melt layer after it froze, it appears likely that it could itself frequently transform into faceted crystals (K. W. Birkeland, personal communication, 2008) of higher SSA. Faceted crystals have a SSA in the range 8–45 $m^2\,kg^{-1}$ (Domine et al., 2007a) so that we expect this transformation to increase SSA, in a manner similar to the case we observed. Birkeland's observations were in Southwestern Montana, but probably apply to many south facing slopes at mid-latitudes.

These Arctic and mid-latitude observations suggest that melt-freeze crusts of low SSA can transform into faceted crystals or depth hoar of higher SSA whenever large temperature gradients (100 to 200°C m^{-1}) are present. This can happen frequently in the Arctic and subarctic due to low temperatures and thin snowpacks, or at mid-latitudes due to diurnal temperature cycling.

Melt-freeze crusts being thin, their SSA increase will have little effect on the snowpack's ability to store adsorbed chemicals. The most notable effect will be the increased permeability, from about $1\times10^{-10}\,m^2$ for refrozen layers (Albert and Perron, 2000) to $500\times10^{-10}\,m^2$ for depth hoar (Shimizu, 1970; Sturm, 1991). This transformation will therefore restore the ability of the underlying layers to exchange chemical compounds with the atmosphere.

Significant radiative effects are expected only if this transformation takes place at or very near the snow surface. We quantified this effect using the DISORT software (Stamnes et al., 1988) which assumes that snow is made of spheres. We used a lognormal size distribution for the spheres, diffuse light conditions, and considered the spectral range 300–2500 nm. Transforming a 5 mm-thick refrozen surface layer of SSA=3.4 $m^2\,kg^{-1}$ into snow having a SSA of 8.8 $m^2\,kg^{-1}$ increases albedo by 1.2%, from 0.776 to 0.788, if the refrozen layer sits above a semi-infinite layer of SSA=8.8 $m^2\,kg^{-1}$.

The radiative forcing caused by this albedo increase is significant. Assuming a daily-averaged incoming solar flux of 200 $W\,m^{-2}$, the forcing at the surface is then 2.4 $W\,m^{-2}$, greater than the 1.66 $W\,m^{-2}$ for the radiative forcing caused by the CO_2 increase between 1750 and 2007 (IPCC, 2007). Furthermore, this 2.4 $W\,m^{-2}$ forcing caused by snow has a stronger climatic effect than the same forcing by CO_2, because its forcing efficacy, i.e. the temperature rise caused by that forcing over the temperature rise due to an equivalent forcing by CO_2, is 3 times as large (Hansen et al., 2005).

4.2 Effect of wind on SSA

Here we find that wind action on Arctic snow that is 2 to 3 weeks old results in a SSA increase, because of a decrease in crystal size due to sublimation and because of fragmentation. Cabanes et al. (2002) observed that wind accelerated SSA decrease of fresh dendritic snow because of the disappearance by sublimation of small microstructures having a high SSA. From these limited observations, we speculate that the vigorous remobilization of snow by wind may form layers of SSA between 40 and 65 $m^2\,kg^{-1}$. Therefore, the SSA of snow with high SSA will decrease, while that of snow with low SSA will increase.

At mid-latitudes where precipitation events are more frequent, we suggest that wind storms will often decrease SSA. In Polar Regions, where precipitation events are infrequent, we speculate that wind action will increase the SSA of surface snow at least in 50% of cases, resulting in albedo increases. This is consistent with Grenfell et al. (1994) and Liljequist (1956), who observed that snow albedo increased after wind storms in Antarctica.

In our example from Alert, we calculate again using DISORT that the replacement of both 1 cm-thick surface layers with a twice as dense 1 cm-thick layer of SSA=61 $m^2\,kg^{-1}$ will increase albedo by 0.9%, from 0.843 to 0.852. Assumptions are as above, and surface layers sit above a windpack of SSA=15 $m^2\,kg^{-1}$. If on the other hand, the new layer covers only 50% of the windpack and is then 2 cm thick, the albedo will decrease by 1.9% to 0.833, because of the lower albedo of the exposed windpack. The radiative and climatic consequences of these effects cannot be neglected, as detailed in the previous section.

Regarding the effect on contaminants, it is clear that SSA changes of 30% in a thin surface layer will not produce major changes in the snowpack loading of adsorbed pollutants. However, for contaminants of moderate to low volatility, most of the contaminant in the (snow+boundary layer) system will be in the snow. Simple calculations, as done by Taillandier et al. (2006) show that increases in SSA from 40 to 60 $m^2\,kg^{-1}$ for a 2 cm-thick snow layer of density 200 $kg\,m^{-3}$ can decrease its boundary layer concentration by 1 to 15%, depending on its volatility.

Wind-pumping of small snow crystals into snow and the resulting SSA increase is a new process that requires additional observations to be confirmed. We also expect this to produce an albedo increase, as well as an enhanced uptake of chemicals.

5 Other considerations

Other processes not observed here could lead to SSA increases. If surface hoar (SSA=24–56 $m^2\,kg^{-1}$, Domine et al., 2007a) forms on an aged wind slab of SSA=15 $m^2\,kg^{-1}$, the SSA of the surface snow will increase. Likewise, fog de-

position, as frequently observed at the top of the Greenland ice cap in summer (Bergin et al., 1995; Domine et al., 1995) might lead to an SSA increase. However, the SSA of deposited fog has not been measured. Diamond dust, i.e. hardly perceptible clear sky precipitation formed of very small crystals that probably have a SSA on the order of 300 $m^2\,kg^{-1}$ (Walden et al., 2003), is also likely to produce an SSA increase, without the appearance of a new snow layer.

Surface hoar formation may explain observations by previous authors. Pirazzini (2004) observed that at Hell's Gate (coastal Antarctica), the snow albedo increased during the night between 1 and 2 December 1997, in the absence of precipitation and under clear sky conditions. Pirazzini does not report any snow observations, but we speculate that this was due to surface hoar deposition on aged snow. McGuffie and Henderson-Sellers (1985, and references therein) also interpret diurnal albedo hysteresis at Resolute (75° N, Canadian Arctic) by the nighttime formation of surface hoar, with daytime sublimation.

In a global warming context, changes in snow albedo can exert powerful forcings and feedbacks (Jacobson, 2004; Hansen et al., 2005; Flanner and Zender, 2005; Flanner et al., 2007). It is therefore essential to understand the causes of observed changes in snow albedo. While the formation of snow crystals of higher SSA is a possibility, another one is the formation of snow with fewer absorbing impurities. For example, the deposition of clean surface hoar of SSA=30 $m^2\,kg^{-1}$ on recent snow of SSA=60 $m^2\,kg^{-1}$ but with a high soot content can increase surface albedo. Changes in snow albedo cannot always be interpreted in terms of snow physics only. If the predictive, physically-based modeling of the evolution of snow albedo is sought, effects on both light scattering and absorption by snow must be understood. We then suggest that snow albedo, snow SSA and concentration of light absorbers be measured simultaneously to reach this understanding. Crystal shapes may also be studied, as these affect albedo (Neshyba et al., 2003; Picard et al., 2009).

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