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Assessing source areas of pollutants from studies of fly ash, charcoal, and pollen from Svalbard snow and ice

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[1] Temporal variations in the quantities of pollen, charcoal, and SCP (spheroidal carbonaceous particles) contained in the Lomonosovfonna ice core from Svalbard and in the surface snow of the ice cap are presented. The abundance of these microparticles is illustrated for the period A.D. 1850–1997 from the ice core, together with detailed snow pit stratigraphies for summer 2001 to spring 2002. Viewing SCP and pollen together is a new approach with relevance for tracing the possible origin of contaminants. The SCP are related to the pollution source while the pollen gives an indication of the vegetation at the point of origin. The deposition of SCP on the ice is also compared with deposition of SCP in northern Fennoscandia for the period A.D. 1982–1997. The quantity of pollen and the pollen types recorded suggest an origin in Fennoscandia rather than in North America. The temporal pattern of both SCP and charcoal deposition reflects that of Europe but may also contains a local signal.

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1. Introduction

[2] The high concentrations of different pollutants, in particular organic compounds, in the pristine Arctic environment and wildlife have received much attention during the last two decades [*Arctic Monitoring and Assessment Programme*, 2002]. One such pollutant, is spheroidal carbonaceous particles (SCP) which, together with inorganic ash spheres (IAS), compose the fly ash produced by burning fossil fuels at high temperatures (see *Rose* [2001] for a summary and definition). Several lake sediment analyses have focused on the SCP component of fly ash in order to study the effect of the post industrial impact on the environment [*Alliskaar*, 2000; *Rose et al.*, 1999; *Wik and Renberg*, 1996]. In these, SCP are specifically extracted, being recognized as porous opaque black spheroids in the size range 5–50 μm. Pollen grains are airborne particles with a similar size range (10–130 μm) which can be expected to be transported in a comparable manner. Airborne particles of this size can also act as transporters of particle-associated contaminants: heavy metals, sulphur and organic components [*Cerceau-Larrival et al.*, 1991]. Pollen has one additional useful feature. Because it can be identified to family, genus or sometimes even species, it can be related back to the vegetation which produced it, which can potentially indicate its source area. Thus, assuming that a suite of airborne particles within the same size range will be transported by the same air masses, investigating SCP and pollen together opens up a new dimension for tracing the origin and dispersal of contaminants.

[3] During the last decades numerous ice core studies from polar ice caps have provided temporal records of pollution in addition to climate. The pollution studies have mostly focused on soluble ions and metals [*Boutron et al.*, 1998; *Mayewski et al.*, 1990]. At the same time pollen in ice cores [*Bourgeois*, 1986] and from pollen traps located on the ice sheets [*Rousseau et al.*, 2003] has been shown to be a useful tracer for major transport paths. Although researchers have looked at the pollen content of ice cores and surface snow [*Bourgeois*, 1990, 2000; *Bourgeois et al.*, 2000, 2001; *McAndrews*, 1984; *Short and Holdsworth*, 1985], other similar-sized airborne particles, in particular SCP and charcoal, have not received the same attention, despite the fact that, from sediment studies, they are known to be as abundant as pollen. In a previous paper we presented some pollen and SCP data from the Lomonosovfonna ice core from Svalbard (Figure 1) [*Isaksson et al.*, 2003]. Here, we extend this series further back in time, integrate the record with snow pit samples from the same snowfield and make a comparison with records of terrestrial SCP deposition monitored by pollen traps in northern Fennoscandia.

[4] Svalbard (Figure 1) is remote from major pollution sources. Considerably smaller sources, but much more local ones, are the coal-fired power stations at Barentsburg, Pyramiden and Longyearbyen (the largest settlement, in operation since 1911). The stations at Barentsburg and Pyramiden, however, ceased operations in 1998 [*Rose et al.*, 2004]. During the winter easterly and southeasterly winds dominate over Svalbard and bring anthropogenic

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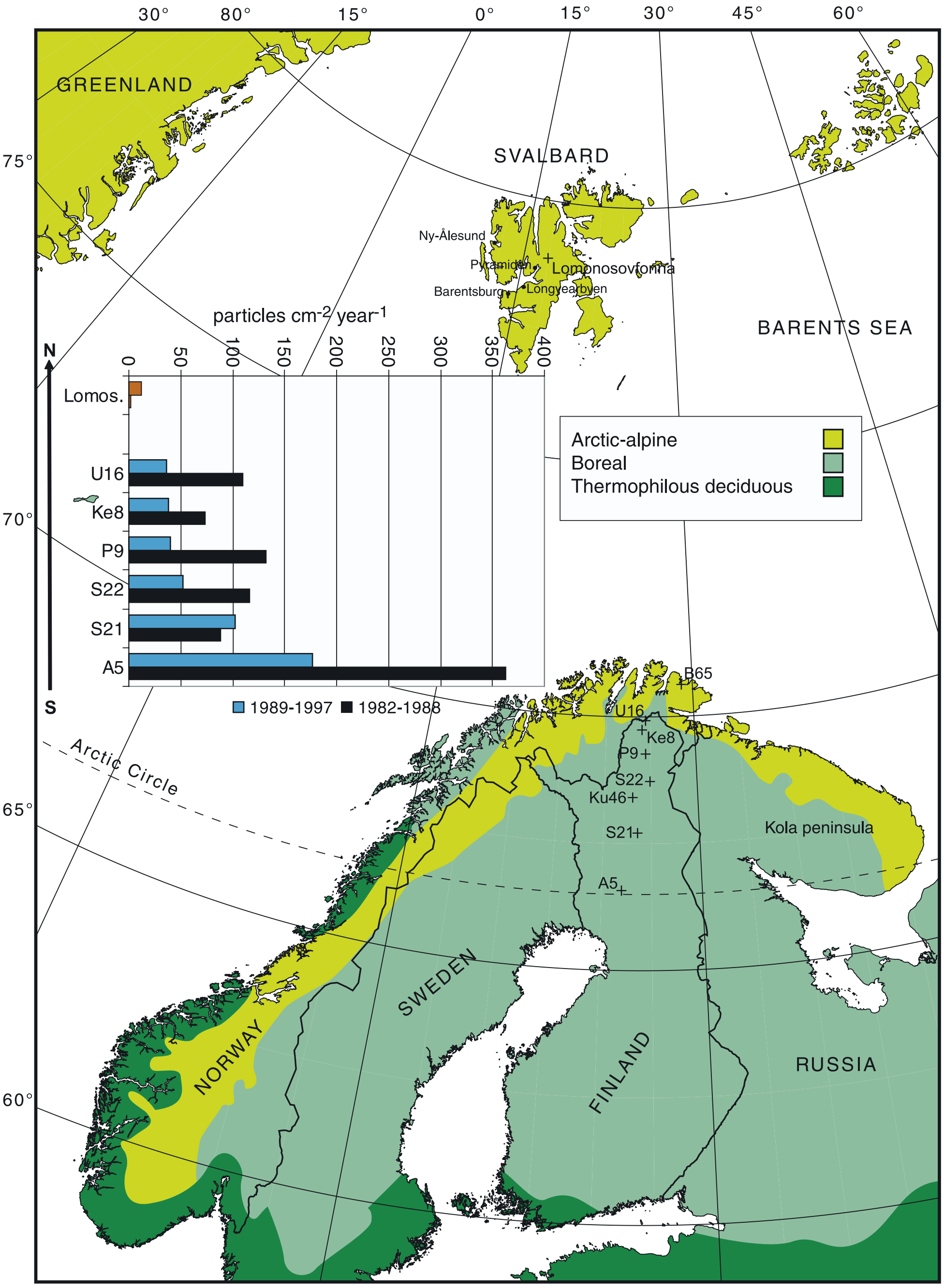

Figure 1. Map showing the location of the major vegetation units in Fennoscandia and the location of the Lomonosovfonna ice core and pollen traps mentioned in the text. The average deposition of spheroidal carbonaceous particles (SCP) in the pollen traps and the ice core (highlighted in red) for the periods 1982–1988 (black) and 1989–1997 (blue) are also shown.

Arctic haze to the islands [e.g., *Heintzenberg*, 1989]. Evaluation of air mass trajectories and the seasonal movement of the atmospheric polar front shows that Svalbard is affected by long-distance transport of contaminants from industrial areas, including eastern and western Europe and Canada [*Eneroth et al.*, 2003]. A model tracer study suggests that European emissions are primarily responsible for the Arctic haze problem [*Stohl et al.*, 2002]. The influence of North Atlantic Oscillation (NAO) on the transport of European pollution has been discussed by [*Eckhardt et al.*, 2003]. They suggested that during the positive NAO phases, which has been strong during the last 2 decades, surface concentrations of trace gases in the Arctic originating in Europe and NW Russia are enhanced by as much as 70% [*Hermanson et al.*, 2005].

2. Methodology

2.1. Ice Core Setting and Dating

[5] The 121 m deep Lomonosovfonna ice core covering about 800 years was drilled in April of 1997 on the highest ice cap in Svalbard 1250 m a.s.l. (Figure 1). The total ice

depth at the drill site was estimated to 123 m using a 50 Mhz ice radar system [*Kekonen et al.*, 2005b]. More details about the site have been described in [*Isaksson et al.*, 2001].

[6] An AWS at the summit reveals that the current temperature range is from 0°C to about −40°C. We do not have any direct observations of the dominating wind direction on the ice cap but on the basis of our work with the chemical records in the ice core it has been suggested that this ice cap is dominated by air masses from the Barents Sea [*Kekonen et al.*, 2005b; *O'Dwyer et al.*, 2000]. On the basis of the concentrations of ions in the snow which are associated with Arctic haze, Lomonosovfonna seems to be positioned above the inversion layer [*Virkkunen*, 2004].

[7] The ice core has been subsampled with 5 cm resolution and analyzed for major soluble ions, ^{2}H and δ^{18}O. The ^{137}Cs content was determined by high-resolution gamma spectrometry and the 1963 radioactive layer was determined to be at 18.50–18.95 m core depth, providing an important reference horizon for dating and estimation of the mean accumulation rate [*Pinglot et al.*, 1999]. For more information on the analytical methods we refer to *Isaksson et al.* [2001] and *Kekonen et al.* [2005b].

[8] The timescale of the core was based on a simple age-depth model based on pure shear according to *Nye* [1963]. The input parameters being the total ice depth at the drill site and the mean accumulation rate tied with the known dates of prominent reference horizons, the 1963 radioactive layer, already mentioned, and the 1783 Laki volcanic acid layer at 66.8 m [*Kekonen et al.*, 2005a]. The Nye model was slightly modified using two different accumulation rates as input parameters [*Kekonen et al.*, 2005b] instead of only one as in the original model by *Nye* [1963]. In addition, the chemical analyses suggest that annual or pseudoannual signals are preserved and this has made it possible to obtain an annual timescale back to at least AD 1715 [*Isaksson et al.*, 2001; *Pohjola et al.*, 2002a]. For the time period before that there is no direct dating, however the δ^{18}O profile from the ice core is in agreement with other available climatic proxies from Svalbard and the Arctic [*Isaksson et al.*, 2003, 2005] and we are thus confident in the timescale.

[9] [*Pohjola et al.*, 2002b] studied the effects of the periodic melt on both the ions and isotopic signals through different quantitative approaches and found that during the warmest summers (such as during the 1990s) as much as 50% of the annual accumulation may melt and percolate down 2–8 years. The summer melt is refrozen within the annual layer however by using multiyear samples, as we have done in this work, this smoothing effect averages out.

2.2. Analytical Methods for Pollen and Fly Ash

[10] From the uppermost 36 m of the core, corresponding to the time between A.D.1900–1997, the ice for pollen, SCP and charcoal samples were taken from 12 contiguous slices where each sample is estimated to cover about 7 years [*Isaksson et al.*, 2003]. Below this depth seven samples were extracted from different depths each covering approximately a 10 year period in the following time ranges: A.D. 1400–1450, 1450–1500, 1850–1859, 1860–1869, 1870–1879, 1880–1889 and 1890–1899, respectively.

[11] At the ice core site on Lomonosovfonna two snow pits 1.4 m (pit 2) and 1.2 m (pit 4) deep were dug in April 2002. From the walls of these pits a series of blocks were cut such that they had a constant surface area and formed a consecutive sequence down the pit wall. In pit 2 they were 15 cm in thickness in the upper part of the pit, but thinner at greater depth. In pit 2 the summer layer was visible as a very distinct ice layer after a particularly warm summer and this was sampled separately. The blocks from pit 4 were thicker and only three samples were taken. Dating of the snow pits was done using a combination of δ^{18}O and major ions [*Virkkunen*, 2004].

[12] Both the snow and ice core samples were melted in closed glass beakers and a tablet containing a known quantity of an 'exotic marker' (*Lycopodium* spores, [*Stockmarr*, 1971]) was added to each sample after melting. The sample was then filtered, and the filter paper acetolyzed, the residue treated with 10% KOH, washed and mounted in silicone oil. From this a subsample was taken for microscope analysis. During the analysis (at magnification of ×400) all pollen grains and spores encountered along the microscope traverses were identified and counted, together with two size classes of charcoal (<40 μm and >40 μm), SCP and the added *Lycopodium* markers. Pollen and spores (including both bryophyte and fungal spores) were identified with reference to the works of [*Faegri and Iversen*, 1989; *Moore et al.*, 1991; *Reille*, 1992; *Van Geel et al.*, 2003] and to the reference collection housed at the Institute of Geosciences, University of Oulu, Finland. Pollen morphological nomenclature follows that of the EPD (European Pollen Database). Algae were also identified from the same preparations [*Jankovska and Komárek*, 2000]. On the pollen slides SCP were recognized as black spheroids of varying size and charcoal as black straight-edged fragments. Since the SCP were mostly at the smaller end of the size range it is possible that the records also include some black-colored IAS, which would indicate coal rather than oil combustion as the source. A more problematical misidentification could be with the conidia of *Nigrospora* spp. which are also black and globose and fall within the same size range. Although such conidia have been isolated from goose dung on Svalbard, they are not among the commonly found dung palynomorphs encountered in the course of pollen analysis [*Van Geel et al.*, 2003] and their shape is slightly ellipsoid. We believe that the spheroids encountered were, indeed, fly ash.

[13] The amount of pollen, SCP and charcoal present in each sample is calculated relative to the added *Lycopodium* marker. As we have reported before [*Isaksson et al.*, 2003], the amount of pollen recovered from the snow pit and ice core samples is insufficient for any sort of statistical analysis. For this reason we have chosen to express the results as grains/particles/fragments cm^{-2} yr^{-1} for the ice core and grains/particles/fragments per liter of water for the snow pit (Figure 2). In order to convert the snow samples to liters of water we used the weight of the samples thus taken varying density into account. The number of added marker grains greatly exceed the amount of counted pollen (which was sometimes less than 10 grains per preparation) so that the confidence limits on the calculated pollen and SCP accumulation rates are minimized [*Maher*, 1981]. We feel, therefore, that these values are more meaningful than percentage ones would be.

[14] The pollen, spores and other microfossils are grouped into three categories on the basis of their probable

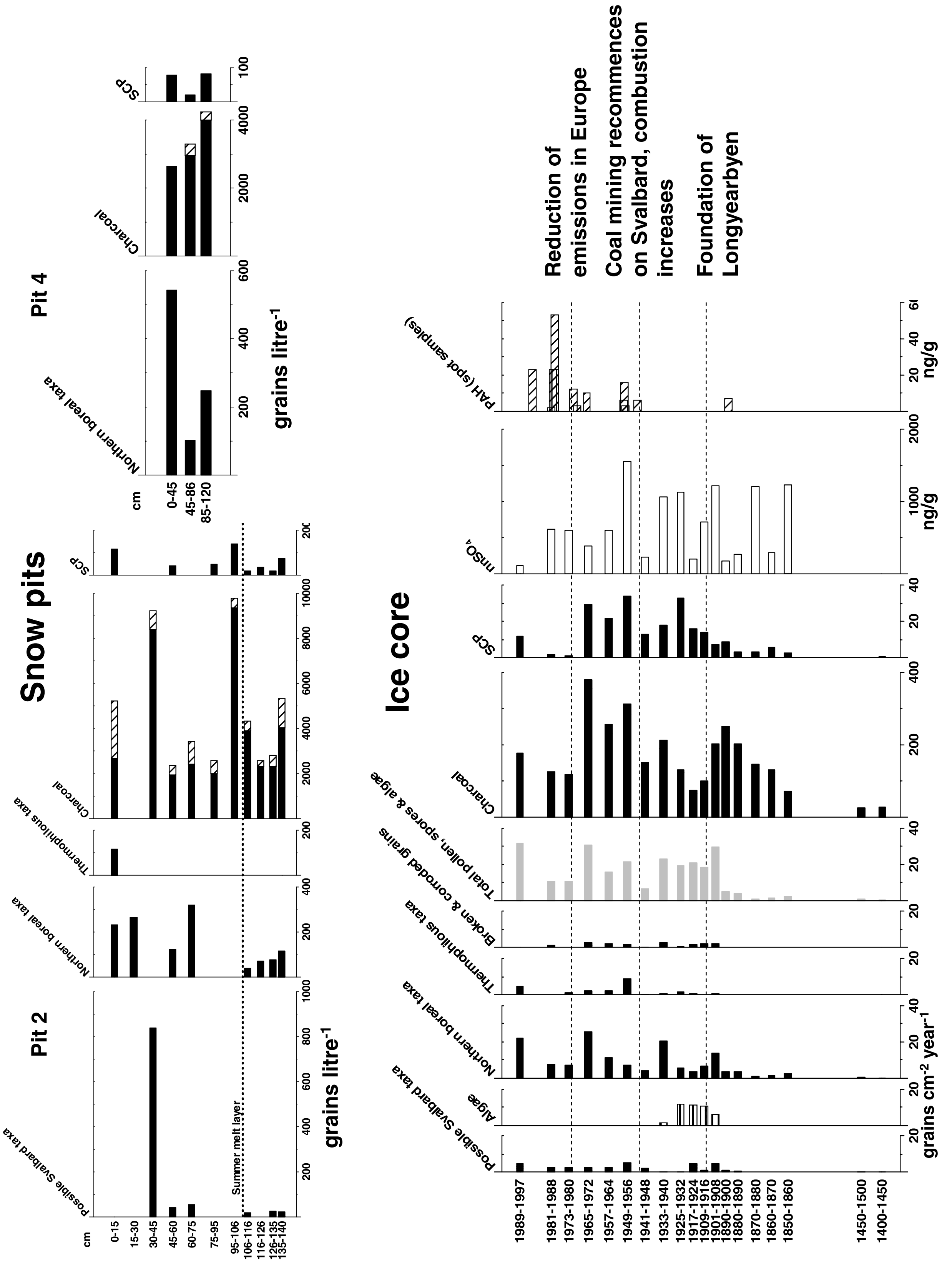
Snow pits
Pit 2
Pit 4
Ice core
Possible Svalbard taxa
Northern boreal taxa
Thermophilous taxa
Charcoal
SCP
Algae
Broken & corroded grains
Total pollen, spores & algae
nnSO4
PAH (spot samples)
Summer melt layer
grains litre-1
grains cm-2 year-1
ng/g
cm
Reduction of emissions in Europe
Coal mining recommences on Svalbard, combustion increases
Foundation of Longyearbyen

Figure 2

Table 1. Allocation of Pollen, Spore, and Algal Taxa to the Three Groups: Possible Svalbard Taxa, Northern Boreal Taxa, and Thermophilous Taxa

	Possible Svalbard Taxa	Northern Boreal Taxa	Thermophilous Taxa
Trees		*Alnus, Betula pubescens* type, *Betula tortuosa* type, *Picea, Pinus, Populus, Sorbus*	*Acer, Corylus, Fagus, Fraxinus, Quercus, Tilia, Ulmus*
Shrubs		*Juniperus*	
Dwarf shrubs	*Betula nana, Salix*	*Vaccinium*	
Herbs	*Cerastium,* Chicoriaceae undiff., *Chrysosplenium,* Cruciferae, Cyperaceae, *Dryas, Equisetum,* Gramineae, *Potentilla,* Ranunculaceae, *Sagina type, Silene dioica*	*Artemisia,* Cannabaceae, *Carduus, Centaurea cyanus, Cerealia, Filipendula, Galium,* Lamiaceae, *Parnassia palustris, Peucadanum type, Prunella,* Rosaceae, *Scheuchzeria,* Scrophulariaceae, *Solidago, Thalictrum, Trientalis, Typha latifolium,* Umbelliferae, *Urtica, Veronica*	*Conium, Helianthemum, Heracleum, Impatiens, Lycopus*
Club mosses		*Lycopodiaceae, Lycopodium annotiunum*	
Ferns and mosses	Polypodiaceae, *Sphagnum*		
Algae	*Botryococcus, Pediastrum*		
Aquatics		*Isoetes*	

source of origin: (1) possible Svalbard taxa, pollen and spores which could come from locally growing plants and locally occurring algae; (2) northern boreal taxa, pollen and spores from plants commonly present in northern Fennoscandia; and (3) thermophilous taxa, mostly (though not entirely) with a more distant source than the northern boreal taxa (Table 1).

[15] A network of modified Tauber-type pollen traps has been monitoring pollen and spore deposition in northern Fennoscandia since 1982. Other microfossils (testate amoebae and stomata), together with charcoal and SCP are also analyzed from the same samples. Details of the methodology and laboratory preparation for these samples are given by *Hicks* [2001]. Annual SCP deposition records (1982–2002) from seven pollen traps on a north – south transect in eastern Fennoscandia are used here (Figures 1 and 3; see also *Hicks* [2001] for their exact locations).

3. Results and Discussion

[16] Pollen and/or charcoal were present in all levels (i.e., seasons) in both of the snow pits but in very small quantities and in all except one layer (30–45 cm in pit 2), northern boreal taxa were obviously more abundant than local taxa (Figure 2). In this layer, however, the pollen type is *Betula nana* type, which could as well originate from the northern boreal forests. The quantities of pollen, up to 400 grains L^{-1} (two samples somewhat more), are comparable to those from the Devon, Penny and Academy of Sciences ice caps but considerably higher than for the other ice caps (Table 2). Since the uppermost six sample layers in pit 2 and all three layers samples in pit 4 cover the winter months it can be concluded that pollen, charcoal and SCP are all present in the atmosphere during the whole year. We believe that these particles are found in the horizons in which they were deposited. The blackness of the SCP and the charcoal would mean that they are good absorbers of radiation but not in sufficient amounts to melt the ice and allow them to move through the snow layers. If running water was present some percolation with gravity could take place but would not penetrate the summer surface ice level. The fact that particles were found more abundantly above this level indicates that no large-scale percolation had taken place. Since the late 1990s summer temperatures have been some of the warmest in the nearly 90 year long record (http://met.no/met/ver_100/klimahistorie/99841_sommar.jpg). On the basis of this and the fact that the $\delta^{18}O$ record [*Isaksson et al.*, 2003] suggests that the 1900s have been the warmest century since the 1400s we assume that the effect of melt has been less down core than might be deduced from the snow pits.

[17] It is to be expected that SCP are emitted to the atmosphere all year round possibly even more abundantly in winter than in summer. Pollen is naturally more abundant during the summer at the time of flowering but can be refloated and recirculated during the winter. Earlier work [*Hicks*, 1985] has confirmed the presence of pollen in the atmosphere over land during the winter, while studies of colored winter snow in Scandinavia [*Franzen and Hjelmroos*, 1988] and experiments testing pollen deposition during the winter [*Hjelmroos*, 1996] have demonstrated that refloated pollen is present in the atmosphere at that time of year and can be transported over considerable distances. Although these factors do not automatically indicate a circum-Arctic distribution, they could be in agreement with such a situation. The ice core samples have a larger than normal percentage of

Figure 2. Pollen, charcoal, SCP (partly adapted from *Isaksson et al.* [2003] and extended with extra samples), non-sea-salt sulphate ($nssSO_4$) [*Kekonen et al.*, 2005b], and Polycyclic Aromatic Hydrocarbons (PAH) [*Vehviläinen et al.*, 2002] records from the Lomonosovfonna ice core together with pollen, SCP, and charcoal records from the snow pit layers sampled in April 2002. A composite record of all the palynomorphs counted (including algae) is additionally included (shaded). The diagonally hatched area in the charcoal records show fragments >40 μm in size. The pollen charcoal and SCP in the ice core are expressed as grains/fragments/spheroids cm^{-2} yr^{-1}, the PAH (finely hatched bars) and the SO_4 (open bars) as ng/g. The pollen charcoal and SCP in the winter snow is expressed as gains/fragments/spheroids L^{-1} of water.

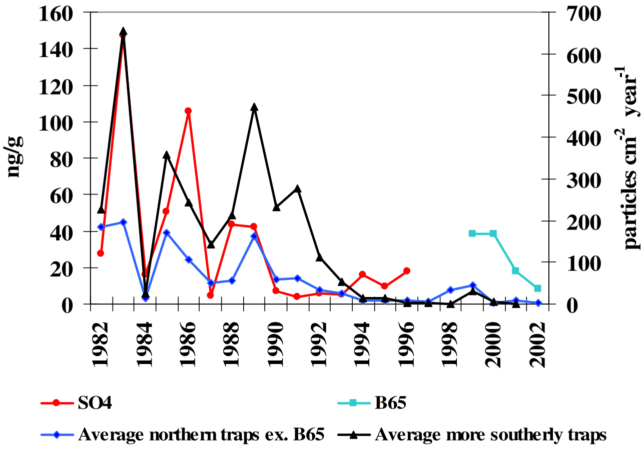

Figure 3. Annual variation in SCP deposition, average separately for the three northern (U16, Ke8, and P9, in blue) and the three more southerly (S22, S21, and A5, in black) pollen monitoring sites for the period 1982–2002 compared with the SO_4 (red) content of the ice core. The shorter record from monitoring site B65 (pale blue) is also included.

corroded and broken pollen grains (compared with the situation in the terrestrial pollen traps) suggesting that the pollen may be redeposited or recirculated and so may have spent a longer time in the atmosphere than locally deposited pollen.

[18] For the ice core samples, where an age depth chronology is available and the volume of each sample is known, it was possible to express the pollen content as pollen accumulation rates (PARs), i.e., grains cm^{-2} yr^{-1}. For the other ice caps the pollen quantity is mostly expressed as grains L^{-1} but in those instances where PARs have been calculated (Table 2) the quantities of pollen are generally less than those recorded for Lomonosovfonna.

[19] We use the probable origin of the different pollen groups to provide information on dominating air masses. Pollen of northern boreal and thermophilous taxa are more abundant than pollen of local Svalbard taxa (this latter group includes not only herbs but also algae) which confirms air mass movement over long distances. However, there are no pollen taxa present which would indicate that the origin of the air masses was in North America, rather than Europe, in contrast to records from the Greenland [*Rousseau et al.*, 2003], Penny [*Short and Holdsworth*, 1985] and Agassiz [*Bourgeois*, 2000] ice caps where north American taxa such as *Ambrosia, Nyssa, Carya* and *Tsuga* are commonly recorded.

[20] The oldest parts of the ice core (14th and 15th century) contained very little pollen. Possible Svalbard taxa are clearly more abundant during the period 1901–1932, although closer inspection shows that these are primarily the green algae; *Botryococcus* and *Pediastrum,* rather than higher plants. Although these taxa do not grow on the ice they are found as communities in Arctic water bodies so, if locally wind transported, are likely to be numerously present. The amounts of both northern boreal and thermophilous taxa appear to be higher in the period 1949–1997 than during the beginning of the century which could possibly indicate a change in the major transport direction or stronger winds in the second half of the century. Broken and corroded grains occur in constant quantities throughout supporting the suggestion that a large proportion of the pollen has been redeposited or recirculated and may have been in the atmosphere for a considerable time.

Table 2. Summary of the Number of Pollen grains L^{-1} Snow/Ice and/or Grains cm^{-2} yr^{-1} as Others Have Recorded for Different Parts of the Arctic[a]

Location	Time Period	Pollen Quantity, grains L^{-1}	Pollen Quantity, grains cm^{-2} yr^{-1}	Source
Agassiz ice cap	1982–1989	1.4–87		*Bourgeois* [1990]
Agassiz ice cap	1900–1950	15		*Bourgeois et al.* [2000]
Agassiz ice cap	last 1000 years	4–22		*Bourgeois et al.* [2000]
Agassiz ice cap 1	1981–1994	2–90		*Bourgeois* [2000]
Agassiz ice cap 2	1990–1994	5–20		*Bourgeois* [2000]
Quviagivaa Glacier	1993	50–345		*Bourgeois* [2000]
Devon ice cap	1989–1994	up to 400		*Bourgeois* [2000]
Devon ice cap	whole core	1.7–13		Litchi-Federovich (1974)[b]
Devon Island	late Holocene	av. 7.4		*McAndrews* [1984]
Academy of Sciences ice cap	1991–1992	5–598		*Bourgeois* [2000]
Penny ice cap	1992 and 1995	4–424		*Bourgeois* [2000]
Penny ice cap	whole core	41–124		*Short and Holdsworth* [1985]
Penny ice cap	1971–1979		1–8	*Short and Holdsworth* [1985]
Greenland ice cap	whole core	21–96		Fredskild and Wagner (1974)[b]
Arctic Ocean	1982–1990	1–17		*Bourgeois et al.* [2001]
High Arctic	1984–1993	5–47		*Bourgeois et al.* [2001][c]
Mid-Arctic	1992	36–43		*Bourgeois et al.* [2001][c]
Low Arctic	1983	17–195		*Bourgeois et al.* [2001][c]
Lomonosovfonna ice cap	1850–1900	41–150	1–5	this paper
Lomonosovfonna ice cap	1909–1948	202–964	11–32	this paper
Lomonosovfonna ice cap	1949–1997	350–921	7–23	this paper
Lomonosovfonna snow pits 2 and 4	2001–2002	50–850		this paper

[a]The Lomonosovfonna records are set alongside these for comparison. For the latter the values given in Figure 2 are averaged for three periods, each covering c. 50 years. Values for the two snow pits are also averaged.
[b]Quoted by *Short and Holdsworth* [1985].
[c]Records from land not included.

[21] Charcoal, particularly the small size class (<40 μm) was the most abundant particle in both the snow and ice samples. Larger pieces of charcoal (>40 μm) and the SCP occurred in frequencies of the same order as the pollen grains (Figure 2). The ice core record suggests that the highest values of charcoal are recorded in the period 1949–1980. Since driftwood would be the only wood source available on Svalbard for local domestic burning, we assume that much of the charcoal must have a more distant origin, most probably forest fires within the boreal zone.

[22] The SCP record shows a rise beginning at the end of the 19th century, in keeping with the expansion of industry in Europe. Rising values at the very beginning of the 20th century are also concurrent with the foundation of Longyearbyen and the beginning of coal mining. Coal mining itself does not produce SCP, however, since the majority of the coal mined went toward power production at the three coal-fired power stations of Longyearbyen, Barentsburg and Pyramiden, in the absence of coal consumption statistics for the first half of the century, coal production provides as useful surrogate [*Rose et al.*, 2004]. Both local and long-distance sources therefore need to be considered. The highest SCP values are seen between 1925 and 1970s, (although there is a reduction in 1930s and 1940s, at the time of the 2nd World War), after which they fall dramatically, in keeping with the laws enforcing a reduction in emissions but rise again slightly in the period 1989–1997. For the period 1970–1995 the temporal distribution pattern of SCP in Lomonosovfonna (Figure 2) is similar to that of some of the lakes in northwest Spitsbergen, with low values in the early 1980s and higher values in the mid 1990s [*Rose et al.*, 2004]. On the longer timescale (from mid 19th century) the temporal pattern on SCP compares with that in varved lake sediments in Sweden [*Wik and Renberg*, 1996]. The quantity of fly ash at Lomonosovfvonna, however, is clearly less than that being deposited in northern Fennoscandia 1–10 particles cm^{-2} yr^{-1} for the period 1981–1997 compared with an average of 50–80 particles cm^{-2} yr^{-1} in the northernmost Finnish sites for the same period (traps U16, Ke8, P9, S22 and Ku46, Figure 1). Only one of the Spitsbergen lakes analyzed by *Rose et al.* [2004] has calculated SCP accumulation rates; Arresjøen in the far north west of the island [*Rose et al.*, 1999]. Here the value is 1.3 cm^{-2} yr^{-1}, which is of the same order as at Lomonosofvonna. The pollen trap records of SCP deposition show the highest quantities in the southernmost localities (traps A5 and S21: closest to industrial sources Figure 3) but they also show a clear fall in quantity from 1989 onward.

[23] *Rose et al.* [2004] correlate the mid 1990s SCP peak on Svalbard with local coal combustion since ‘maximum total consumption of coal for the Isfjord power stations occurred in 1993’, and conclude that a local contamination signal (recorded over an area 60–80 km from source) is superimposed on the long-range signal. Lomonosovfonna is within this 60–80 km range of the power stations and it is possible that the slightly elevated values of SCP recorded in the 1989–1997 sample could also reflect this peak in local coal consumption (but see also following). This might also be the case with the drop in SCP during the 2nd World War since coal production, and therefore also coal consumption, ceased at that time [*Rose et al.*, 2004]. The ice core record, however, goes back much further than all but one (Tenndammen) of the lake sediment records presented by Rose and the temporal resolution is generally better. Because of the dominance of easterly and southeasterly winds during the winter (the time of maximum power production) it can also be expected that the ice core would be less likely to receive locally produced SCP than the lakes.

[24] At the atmospheric sampling station at Zeppelinfjellet in Ny-Ålesund, 100 km from Lomonosovfonna, black carbon, sulphur dioxide and sulphate concentrations are highly correlated [*Eleftheriadis et al.*, 2001]. SCP form a component of black carbon but not all combustion processes which produce black carbon (e.g., diesel-fired generators as at Ny-Ålesund) produce SCP. It is, nevertheless, interesting to compare the SCP records with the available pollutant records from the Lomonosovfonna ice core. In the Svalbard ice cores anthropogenic influence is illustrated by a rapid increase in sulphate and nitrate levels and in acidity beginning in the late 1940s and continuing to the 1980s, with some decrease in the most recent years [*Isaksson et al.*, 2001]. However, we already observe an increase in non-sea-salt sulphate concentrations in the late 1800s [*Kekonen et al.*, 2005b], in keeping with the rise in SCP. A naphthalene, a polyaromatic hydrocarbon (PAH) component, record from the same ice core suggests that prior to the 1930s naphthalene concentrations were below the determination limit and increased in the 1980s, much in line with the sulphate record [*Vehviläinen et al.*, 2002] (Figure 2). Furthermore correlation of PAH with physical and chemical parameters in the core strongly suggests wintertime deposition of PAH with Arctic haze. The contribution of the coal combustion on Svalbard to the record appears to be small compared with anthropogenic emissions from long-range sources [*Vehviläinen et al.*, 2002].

[25] For the short period for which the pollen trap and the ice core records overlap and can be compared on a year to year basis (1982–1997), the SCP trend is not unlike that of the sulphur emissions (Figure 3). This is to be expected in that both originate from combustion of fossil fuels. It is tempting to suggest that the location of their origin is also the same but the data are insufficient to confirm this. In recent years (1999–2003), SCP deposition is consistently higher at Båtsfjord in northeast Norway (trap B65, Figure 3) than at any of the Finnish monitoring sites, suggesting a different origin, possibly in the Kola peninsula. The higher SCP values at Lomonosovfonna in the period 1989–1997 relative to the period 1982–1988 (the reverse of the Finnish pollen trap records, Figure 1) has been attributed above to local coal combustion (following the observations of *Rose et al.* [2004]) but could also have a more eastern origin following the Båtsfjord record. Unfortunately this record is too short to allow comparison with the early 1980s when SCP values in the Finnish traps were noticeably higher.

4. Summary and Conclusions

[26] The analysis of SCP charcoal and pollen and spores from snow pits and an ice core at Lomonosovfonna and from pollen traps in northern Fennoscandia allows transport of these particles to be assessed at different temporal scales. The snow pits show summer – winter variation, the pollen

traps annual variation and the ice core long-term average annual accumulation. Whereas SCP may be more abundant in the atmosphere during the winter months, charcoal is present all the year round and pollen is most abundant during the summer. Refloatation of pollen, however, means that some is still present in the atmosphere during the winter. In terms of origin, SCP are produced on Svalbard but in relatively smaller quantities when compared with the main industrial centers of the world. Charcoal, in contrast, is less likely to be produced locally because of the lack of wood. The pollen can be attributed to different vegetation zones and therefore roughly separated into possible local taxa, northern boreal taxa and thermophilous taxa.

[27] When these temporal and spatial aspects are combined we come to the following conclusions. Charcoal, SCP, northern boreal and thermophilous pollen taxa are more abundant in the winter layers of snow pits than in the summer. Since it has been shown that the easterly and southeasterly winds dominate over Svalbard during the winter and bring anthropogenic Arctic haze to the islands and since the correlation of PAH with physical and chemical parameters in the ice core strongly suggests wintertime deposition of PAH with Arctic haze, we suggest that the majority of the charcoal, SCP and pollen is also transported by the winter winds. We see the main source as being in Europe.

[28] At the annual scale the similarity between the SO_4 values at Lomonosovfonna and the SCP deposition in northern Finland show that they could have a common source. This would be in keeping with the conclusion based on the winter snow layers. This degree of temporal resolution is lost for the SCP, charcoal and pollen and spore record from the ice core (though not for the SO_4 and PAH) as it is in the lake sediment records. At this coarser temporal resolution long-term average changes are reflected. The presence of broken and corroded grains throughout the ice core, suggesting that these grains have been refloated and recirculated, would be in keeping with our suggestion that much transport is taking place during the winter months.

[29] The long-term average record for the SCP has features which match both the European record and the local Svalbard record. In the light of the other evidence, however, we suggest that the European source is dominating over the local one.

[30] Charcoal and SCP have not been analyzed from the other ice caps so a comparison can only be made with the pollen record. The quantities of pollen recorded at Lomonosovfonna are of the same order, or slightly higher than those recorded in other Arctic ice caps and the pollen types are similarly Arctic-alpine and boreal. The higher values are in keeping with Lomonosovfonna being closer to Fennoscandia and to forest covered areas than the American ice caps are.

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