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Observed metre scale horizontal variability of elemental carbon in surface snow

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Abstract

Surface snow investigated for its elemental carbon (EC) concentration, based on a thermal–optical method, at two different sites during winter and spring of 2010 demonstrates metre scale horizontal variability in concentration. Based on the two sites sampled, a clean and a polluted site, the clean site (Arctic Finland) presents the greatest variability. In side-by-side ratios between neighbouring samples, 5 m apart, a ratio of around two was observed for the clean site. The median for the polluted site had a ratio of 1.2 between neighbouring samples. The results suggest that regions exposed to snowdrift may be more sensitive to horizontal variability in EC concentration. Furthermore, these results highlight the importance of carefully choosing sampling sites and timing, as each parameter will have some effect on EC variability. They also emphasize the importance of gathering multiple samples from a site to obtain a representative value for the area.

Keywords: black carbon, elemental carbon, surface snow, metre scale variability

1. Introduction

Cryospheric changes can provide good indications of climatic change (e.g. Lemke *et al* 2007). Examples of these are observations in changing sea ice cover, glacier mass balances, or the onset of snowmelt. It has been understood for some time that dark impurities in snow can potentially have large feedback effects on the rate of these changes (Warren and Wiscombe 1980, Clarke and Noone 1985, Hansen and Nazarenko 2004).

In the recent decade, black carbon (BC), has been a research area of increasing interest, especially its climatic impact on snow and ice albedo (e.g. Hansen and Nazarenko 2004, Flanner *et al* 2007, McConnell *et al* 2007, Xu *et al* 2009), but also its negative effects on human health (e.g. Anenberg *et al* 2010, Shindell *et al* 2012). Combustion from biomass and fossil fuels are sources of BC particles, hence BC may have both a natural and anthropogenic component.

In this study we will focus on rather small, metre scale, variability of elemental carbon (EC) concentrations in surface snow. For our snow impurity study here, the terms EC and BC are used synonymously, as EC is operationally defined by using a thermal–optical method, while BC and refractory black carbon (rBC) in snow are determined with an optical and a laser-induced incandescence method, respectively. Previous surveys of BC and EC in snow that mainly focused on regional



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(Forsström *et al* 2009, Doherty *et al* 2010, Hadley *et al* 2010, Ye *et al* 2012) and local (Aamaas *et al* 2011) variability also briefly examined small scale horizontal variations in surface snow.

Here, two sites with very different ambient conditions are compared, representing a clean and a polluted environment (Pallas, northern Finland $\sim 68^\circ\text{N}$ and Stockholm, Sweden $\sim 59^\circ\text{N}$). The comparison was conducted on observations made at two different times during the same snow season. The aim of this letter is to investigate the spatial variability of EC on a metre scale in surface snow (top 5 cm) from these different sites, and to analyse possible reasons for this variability.

2. Experiment

2.1. Measurement sites and analytical method

Investigations were carried out at two different sites in the winter and spring of 2010. Tyresta National Park (N $59^\circ 11' E 18^\circ 18'$), circa 25 km from the city centre of Stockholm, Sweden served as the polluted site. Its proximity to an urban area with a population of about 2 million people (metropolitan area), makes the site considerably affected by air pollution. The sampling site in Tyresta, which is an open section of a mire, is exposed to only limited local emissions within about a 5 km radius. Pallas-Yllästunturi National Park (N $67^\circ 58' E 24^\circ 06'$; 450 m a.s.l.), located in Arctic Finland, served as the clean site with no major city influencing the local and regional air.

Samples were collected on Sammaltunturi mountain slightly above the tree line where the terrain was quite flat. Samples from both sites were gathered during winter conditions, on 17 February in Tyresta (hereafter called TY1) and on 3 and 4 March in Pallas (PA1a and PA1b). Supplementary samples during the melting season in spring were collected on 21 March in Tyresta (TY2) and 7 May in Pallas (PA2). The sample strategy consisted of collecting individual surface snow (approximately the upper 5 cm) samples in different grids, with the most common grid-net consisting of a 20 m \times 20 m square with samples taken every 5 m within the square (see figures 1(a)–(e)). Typical sample volumes (melted snow) were around 1 l for both sites.

Snow samples were melted in a glass jar using a microwave oven and filtered through a microquartz filter as in Forsström *et al* (2009) and Aamaas *et al* (2011). The dried microquartz filters containing the collected impurities were analysed in a thermal–optical carbon aerosol analyzer (Sunset Laboratory Inc., Forest Grove, USA) following the NIOSH 5040 protocol (Birch 2003) (see Birch and Cary (1996) for a detailed description of the thermal–optical method). In short, the thermal–optical carbon aerosol analyser first heats the filter (a 1.5 cm² punch) in a helium atmosphere, and organic carbon (OC) and carbonate carbon (CC) are released from the filter and detected by a flame ionization detector (FID). In a second step, the filter is again heated, but in an atmosphere containing oxygen. In the second step, EC is released. However, pyrolysis of the OC which occurred in the

first step is detected as EC in the second step. To account for this, a laser is used to monitor the transmittance of the filter as the transmittance decreases during pyrolysis. The point in time when the transmittance returns to its original value during the second step determines the partitioning between OC and EC in the thermogram.

2.2. Measurement uncertainties

The NIOSH 5040 (Birch 2003) analysis protocol has been argued to underestimate EC content during analysis (Chow *et al* 2001, Reisinger *et al* 2008). Mineral oxides existing on the filter sample can produce oxygen during the first—oxygen free—stage of the analysis. Some of the EC is then incorrectly accounted for as OC during the second stage. In a recent study by Wang *et al* (2012), where they investigated the influence of dust on EC measurements in ice and snow using a thermal–optical method, similar results with a bias in the OC/EC split were found. Chow *et al* (2001) compared the NIOSH protocol with a different analysis protocol, IMPROVE (Chow *et al* 1993), on air samples and found that NIOSH values were typically less than half of IMPROVE values. Similarly, Aamaas *et al* (2011) examined the IMPROVE/NIOSH 5040 ratio on snow samples with the result of IMPROVE having higher EC estimates by a factor of two compared to NIOSH 5040. In this study, an evaluation of the different protocol procedures revealed an average corresponding ratio IMPROVE/NIOSH 5040 that corroborates this approximate factor of two (average values of 2.23, and median value of 1.73, when comparing 10 samples) and as a consequence, the samples analysed with NIOSH 5040 were multiplied by 2 to compensate for the underestimation of the NIOSH protocol.

An assessment of the representativeness of a filter punch (1.5 cm²) for the entire filter (with 11.34 cm² area) was also evaluated. Four punches from each filter, instead of the ordinary one punch, were analysed from a subset of 12 filters (3 from each sampling episode) to observe the representativeness of single filter punches. As expected, greater variability was found for filters with low loading than for filters with high loading. In summary, samples with an EC filter loading of less than $\sim 0.35 \mu\text{g cm}^{-2}$ generated a coefficient of variation (standard deviation as a fraction of the mean, expressed in %) of 40% or greater. Samples with EC filter content greater than $\sim 1 \mu\text{g cm}^{-2}$ produced a coefficient of variation of 23% or less. The average EC content per filter from Pallas and Tyresta was $1.92 \mu\text{g cm}^{-2}$ and $26.6 \mu\text{g cm}^{-2}$, respectively.

A shortcoming of using the filter-based method is that some EC particles can potentially percolate through the filter (Ogren *et al* 1983). This filtration efficiency issue, known as undercatch, was investigated for a subset of filters. Two filter substrates were placed on top of each other in the filter setup, and the sample water was drawn through the filters at the same filtration event. The average and median values from 16 events (12 sub-Arctic snow and 4 Arctic snow samples) were 14% and 2%, respectively. On two additional occasions the melt water was recycled. In this

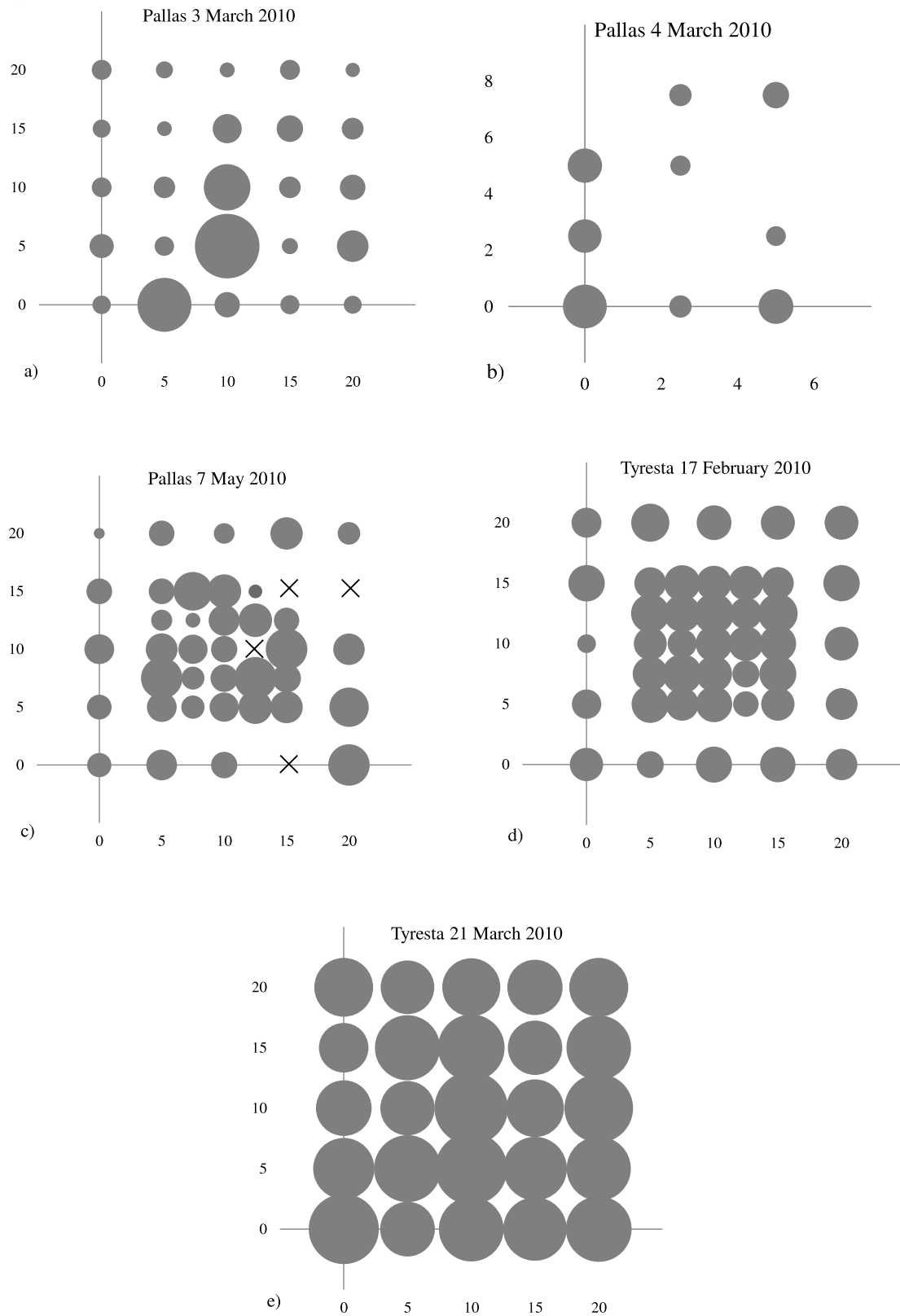


Figure 1. (a)–(e) Circle charts from each sampling event with the area of the circle representing the samples' corresponding EC concentration. The area of the circles are relative as Pallas concentrations are much lower than Tyresta (see table 1). Ordinate and coordinate scales are in metres. Crosses from Pallas 7 May 2010 correspond to samples below the detection limit.

procedure, a sample was filtered once and that collected water was filtered again on another filter. On one of those events, no EC was observed on the second filter and for the

other event, an undercatch of as much as 50% compared to the first filtration was observed. Aamaas *et al* (2011) found similar differences when testing this way. Of three filters, two

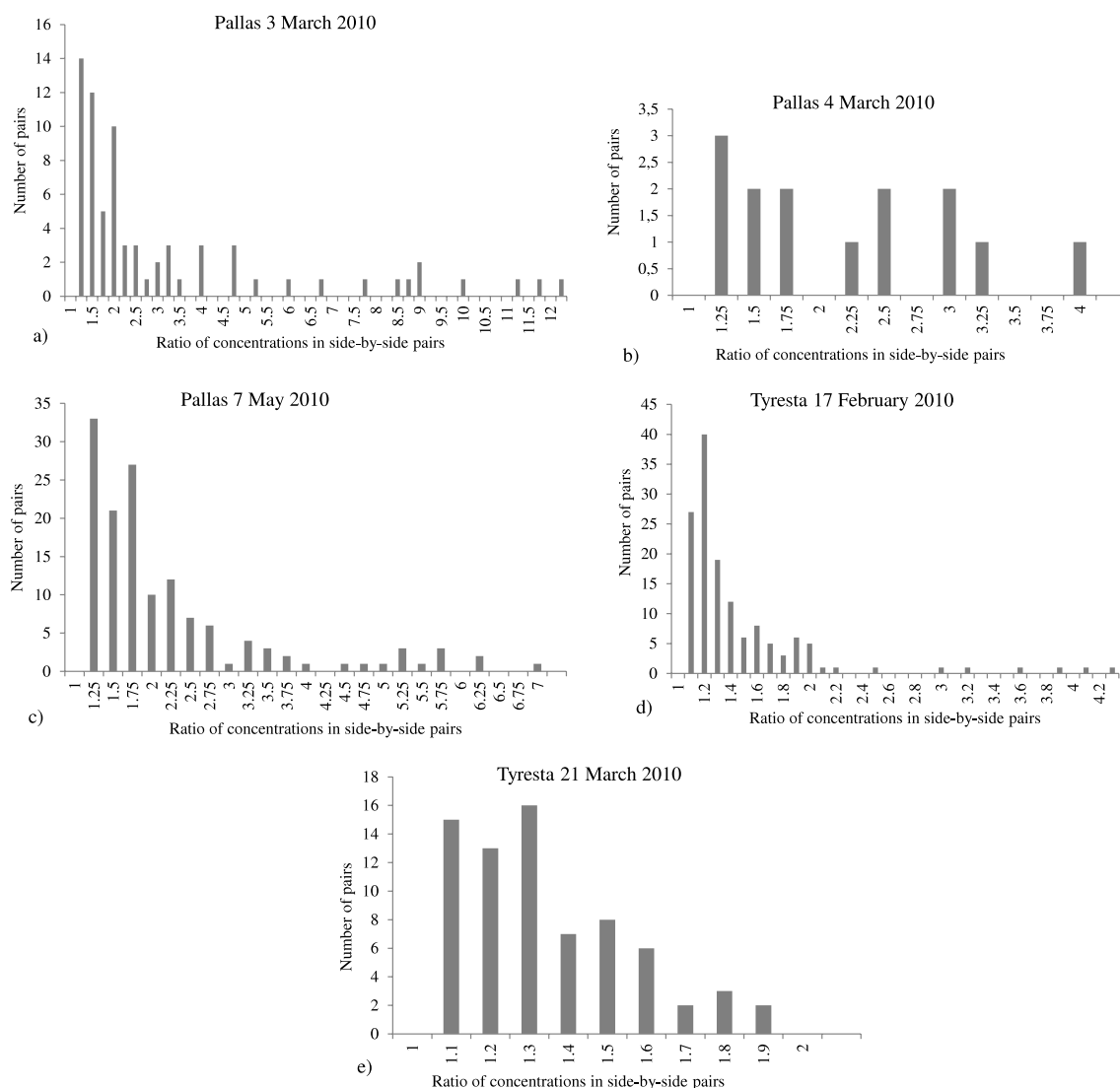


Figure 2. (a)–(e) Side-by-side ratios of neighbouring samples shown in histograms from each sampling event.

had undetectable amounts, while one had a 70% efficiency. These findings demonstrate that undercatch can influence the results and more systematic experiments are needed in the future. Using stacked filters reduces the risk of unwanted loss or contamination compared to recycling the water. For comparison, Nuclepore filters (0.4 μm) used in an optical method using the integrating-sandwich spectrophotometer (Clarke 1982, Grenfell *et al* 2011) have been proposed to have an undercatch of about 15% (Clarke and Noone 1985, Doherty *et al* 2010).

3. Results and discussion

As expected, the typical EC concentration observed at Tyresta was higher than in Pallas. In Tyresta, the measured EC mixing ratio typically ranged from 174 to 505 ppb (25th and 75th percentile, respectively) and in Pallas the range was between 13 and 31 ppb. The concentrations increased from winter to spring at both sampling sites, the median value being roughly threefold from TY1 to TY2 (182.7 versus

608.3 ppb) and twofold from PA1a to PA2 (from 13.0 versus 24.8 ppb). These increases corroborated the accumulation of EC particles occurring throughout the spring in the surface of the snowpack, which has been observed by others (e.g. Conway *et al* 1996, Doherty *et al* 2010, Aamaas *et al* 2011).

Aside from the similarity of a general increase in EC concentrations over the season, other indicators of variability were clearly larger in Pallas than in Tyresta. The variation of EC between samples is illustrated by histograms showing the side-by-side ratio existing between neighbouring samples (distance of 2.5 and 5 m) presented in figures 2(a)–(e). (Ratios presented here are always >1 because the higher concentration is divided by the lower in comparison to Doherty *et al* 2010, figure 6, where ratios below 1 are also presented. This was done here to display a more easily read histogram.) In Tyresta this ratio was typically <2 with a median of 1.2 for all the samples. In Pallas values >2 were common and the median for all samples from there was 1.7. For neither site could the observed variability be explained simply by uncertainties in the sampling procedure,

Table 1. Measured EC concentrations from each sampling event. (Note: coefficient of variation is expressed in percentage.)

Site	Sampling date	Number of samples	Corresponding figures in this letter	Average (ppb)	Median (ppb)	Min (ppb)	Max (ppb)	Standard deviation (ppb)	Coefficient of variation
PA1a	3 March 2010	25	1(a); 2(a)	25	13	6.6	140	30	120
PA1b	4 March 2010	9	1(b); 2(b)	20	15	8.6	42	11	59
PA2	7 May 2010	41	1(c); 2(c)	26	25	0.0	58	16	61
TY1	17 February 2010	41	1(d); 2(d)	180	180	53	250	40	22
TY2	21 March 2010	25	1(e); 2(e)	580	610	370	810	120	20

Table 2. Summary of the side-by-side ratios within each sampling event.

Site	Sampling date	Corresponding figures in this letter	Number of pairs	Max ratio	Average ratio	Median ratio
PA1a	3 March 2010	1(a); 2(a)	72	16	3.2	1.8
PA1b	4 March 2010	1(b); 2(b)	14	3.8	2.1	2.0
PA2	7 May 2010	1(c); 2(c)	140	6.8	2.1	1.6
TY1	17 February 2010	1(d); 2(d)	140	4.3	1.3	1.3
TY2	21 March 2010	1(e); 2(e)	72	1.8	1.4	1.2

reflecting an interesting difference in variability between these two sites. The expected median side-by-side ratio from analysis uncertainty was 1.27, assuming a coefficient of variation of 23% (taken from the representativeness of a filter punch), which is an upper level estimate since the average EC loading is typically higher than for the estimate made here (average loading for Pallas samples was $1.92 \mu\text{g cm}^{-2}$ compared to the $1 \mu\text{g cm}^{-2}$ used for this expected median side-by-side ratio). The samples from PA1a present the largest spatial variability of the data set. This is partially explained by the fact that the three samples with the highest concentrations from all of the Pallas samples were measured at this event (PA1a). The remaining samples at PA1a also differed significantly, indicated by a median side-by-side ratio of 1.81 for all of the samples in this grid-net (table 2). To assess the possibility of contamination in the three samples with the highest concentrations from Pallas (PA1a), the procedure of side-by-side ratios was repeated excluding these three samples. The resulting median side-by-side ratio for the remaining 22 samples in that dataset was 1.54, which demonstrates notable variation regardless of the excluded samples. Although the possibility of contamination cannot be ruled out, contaminating the filters to an EC loading equivalent of the highest concentrations observed requires considerable effort. Simple miss handling of the filter is not sufficient. Hence, it is unlikely that contamination has occurred at Pallas.

A notable variation was also displayed in the grid-net from PA1b, which was a smaller grid-net with a 2.5 m distance between the samples, collected at about the same location, but 24 h after the PA1a event. During the time between the two sampling events, snowdrift had erased all traces of the earlier snow sampling from the previous day. The median side-by-side ratio for this grid-net was 2.0 (figure 2(a), table 2). Wind-driven snowdrift is a common process in Pallas being especially prominent in areas above the tree line. Redistribution of snow from snowdrift has been proposed as an important mechanism creating variations of impurity

concentrations in the snow in other regions of the Arctic (i.e. Svalbard) (Forsström *et al* 2009). The variations observed in Pallas support this view. The snowdrift can mix snow of different ages (with different depositional history) and impurity concentrations, thereby causing particle variability in the snow. This can, in turn, create vertical gradients in the snow that can be translated into horizontal variability when sampling a vertical layer of 5 cm. Doherty *et al* (2010) found that the largest variation between samples were observed in snow pits with strong vertical gradients of BC concentrations. Other recent work on BC in snow, utilizing a different analysis method (the single particle soot photometer), has proposed that there is a significant variability in the size distribution of BC particles in snow (Schwarz *et al* 2012). With the hypothesis that our filter-based method does not capture small EC particles well, as the undercatchment section has suggested potential losses of EC particles while filtering, some of the variation observed in our data could be attributed to the loss of smaller EC particles in our samples. This speculation remains to be further tested, however. It should also be noted that the key result of Schwarz *et al* (2012, 2013) is that BC particles tend to be larger in snow compared to the atmosphere, thus the question of how many small particles are lost in our method is yet to be thoroughly quantified.

The spatial variation in Tyresta was not as pronounced as in Pallas, as reflected by the median side-by-side ratio of 1.25 for TY1 and 1.22 for TY2 (figures 2(d) and (e), table 2). An important element that partially explains the difference in variation between Tyresta and Pallas was the relevant absence of wind and subsequent snowdrift at the former.

The overall variation within the samples from each sampling event was verified by the coefficient of variation from each event (table 1). The sampling events with high coefficients of variation were those from Pallas, especially PA1a (PA1a: 122%; PA1b: 59%; PA2: 61%), whereas the datasets from Tyresta showed much lower coefficients of variation (TY1: 22% and TY2: 20%). The coefficients of variation, together with the side-by-side ratio histograms,

demonstrate the existing variation of EC in the snow from the sites. A recent study conducted in the Sierra Nevada Mountains (CA, USA) by Sterle *et al* (2013) found a spatial variability of rBC in the upper sections of the snow pits during the accumulation season.

The variation presented by Forsström *et al* (2009) had an average relative root mean square deviation of 1.0 on a horizontal scale of 1 m. Thus, the observed variations by Forsström *et al* (2009) share the horizontal variability of EC in snow that is presented here, even though the samples from PA1a, PA1b and PA2 displayed even greater variations than observed by Forsström *et al* (2009). In our case at Pallas, what could set the stage for variability are the existing ambient air conditions. Observations in air have shown polluted air masses originating from Central and Eastern Europe migrating north, while clean air masses from the north also occur at Pallas (Hyvärinen *et al* 2011).

In the work of Doherty *et al* (2010), the variability of BC on a 50–100 cm scale was briefly examined to test the representativeness of a sampling location by individual samples. In a similar side-by-side ratio study carried out with samples from East and West Russia, the Canadian Arctic, and Tromsø, Norway, the concentrations of their samples were almost always within 50% of each other. In fact, most of their samples were typically within 20–30% of each other. Our Pallas samples showed greater variability being >100% from sample to sample. Our Tyresta samples had a variability that was comparable to that presented by Doherty *et al* (2010). Doherty *et al* (2010) does stress the need to gather multiple samples from one location or region in order to attain a representative value, which is also strongly emphasized by the results of this study.

Doherty *et al* (2010) suggested that a closer proximity to emission sources would result in greater depositional heterogeneity of BC. For plumes in the atmosphere this is often the case, but our results indicate that this is not necessary always the case in snow. Tyresta—which is closer to emission sources—showed greater homogeneity than the Pallas samples collected further away from emission sources. The two sites are very different, but post-deposition processes might be more important in a remote environment such as Pallas compared to Tyresta, where stronger dry and wet deposition of EC due to higher ambient air concentrations may help to mask variability caused by snow drift. This would result in a vertically more homogeneous snow pack with respect to EC concentration. This hypothesis remains to be tested.

In light of the results observed in this work, the following recommendations for future sampling of BC/EC in snow are suggested for optimizing measurements: (1) collect multiple vertical profiles a few metres apart to achieve a more representative value for the sampling location, especially during the earlier part of the snow season when the BC loading may be lower; (2) having consistency when sampling layers is of great importance, as well as conducting studies of the layering in the snowpack. Preferably, physical properties of the snow pack (e.g. density, snow grain size, etc) that were not observed in our study, but in light of our results, should be

noted in future studies; (3) when choosing sampling site the local and regional contamination of BC should be taken into account. Areas especially prone to wind exposure should be avoided.

4. Conclusions

Using a comparatively simple filtering technique we observed a spatial variability of EC particles in surface snow on a metre scale. The surface snow displayed greater variability during the winter season and had a tendency towards a more homogeneous pattern during the spring. Greater variability was found for the remote site compared to the site closer to the emission sources.

Our study highlights the importance of carefully choosing the sampling site and timing of the sampling. It also underlines the importance of collecting several samples from a sampling site. EC variation can exist within a few metres in the surface snow. This is also something that needs to be considered when single observations represent large regions in climate models. Our work also argues for the need of studies on EC in snow that focus on both deposition and post-depositional processes taking place in the snow (e.g. microphysics, wind's effect, and metamorphism), as understanding these processes is necessary in order to understand the EC concentration in the snow.

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