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TCD

9, 5053–5095, 2015

A synthetic ice core approach to estimate ion relocation in an ice field site

C. P. Vega et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Abstract

Physical and chemical properties of four different ice cores (LF-97, LF-08, LF-09 and LF-11) drilled at Lomonosovfonna, Svalbard, were compared to investigate the effects of meltwater percolation on the chemical and physical stratigraphy of these records.

5 A *synthetic* ice core approach was employed as reference record to estimate the ionic relocation and meltwater percolation length at this site during the period 2007–2010. Using this method, the ion elution sequence obtained for Lomonosovfonna was $\text{SO}_4^{2-} > \text{NO}_3^- > \text{NH}_4^+ > \text{Mg}^{2+} > \text{Cl}^-$, $\text{K}^+ > \text{Na}^+ > \text{Ca}^{2+}$, with acidic ions being the most mobile within the snowpack. The relocation length of most of the ions was in the order
10 of 1 m, with the exception of SO_4^{2-} showing relocation lengths > 2 m during this period. In addition, by using both a positive degree day (PDD) and a snow-energy model approaches to estimate the percentage of melt at Lomonosovfonna, we have calculated a melt percentage (MP) of the total annual accumulation within the range between 48 and 70 %, for the period between 2007 and 2010 which is above the MP range suggested by the ion relocation evidenced in the LF-syn core (i.e. MP = 30 %). Using a firn-densification model to constrain the melt range, a MP of 30 % was found over the
15 same period which is consistent with the results of the synthetic ice core approach, and a 45 % of melt for the last 60 years. Considering the ionic relocation lengths and annual melt percentages, we estimate that the atmospheric ionic signal remains preserved in
20 recently drilled Lomonosovfonna ice cores at an annual or bi-annual resolution.

1 Introduction

Pollutants produced in low- and mid-latitudes are transported to the polar regions where they are included in the snow by different mechanisms, mainly wet and dry deposition, accumulating in glaciers and ice caps. Major ions, such as SO_4^{2-} , NO_3^- ,
25 Na^+ , and Cl^- , are deposited in the snowpack and can be measured in ice cores, providing valuable information about their sources, chemical transformations in the atmo-

TCD

9, 5053–5095, 2015

A synthetic ice core approach to estimate ion relocation in an ice field site

C. P. Vega et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



sphere, and transport patterns to the sampling site (Laj et al., 1992; Goto-Azuma and Koerner, 2001; Kekonen et al., 2002; Hastings et al., 2004, 2009). Nitrate (NO_3^-), for example, has been used as a record for past atmospheric nitrogen oxides ($\text{NO}_x = \text{NO}_2 + \text{NO}$) (Kekonen et al., 2002; Röthlisberger et al., 2002; Hastings et al., 2005, 2009; Vega et al., 2015b). However, this proxy has been difficult to develop since NO_3^- in snow has several sources and experiences post-depositional processes, such as photolysis, diffusion within the ice, evaporation as HNO_3 , or relocation by meltwater (Goto-Azuma et al., 1994; Honrath et al., 1999; Rempel et al., 2002; Röthlisberger et al., 2002). The latter has been an enigma to potential ice core sites since relocation and preferential elution of chemical species gives an altitudinal and latitudinal threshold for potential drilling sites. To make use of environmental data from outside the dry snow zones on glaciers in the polar areas, it is important to give better regional coverage of the atmospheric chemistry outside the large polar ice sheets. For a correct interpretation of the chemical ice core data from drilling sites where significant summer melting occurs, it is necessary to estimate the post-depositional effects of meltwater percolation on the chemical content of snow and ice.

It is known that most of the ice core drilling sites at Svalbard experience summer melting, which may damp the chemical signal of ionic species, making difficult to understand the transfer function between ion concentrations in the atmosphere and the snow (Goto-Azuma et al., 1994; Isaksson et al., 2001; Moore et al., 2005; Grinsted et al., 2006). Ions do not move uniformly when are removed from the snowpack by meltwater. Instead, they are released following a preferential elution (Goto-Azuma et al., 1994; Eichler et al., 2001; Ginot et al., 2010). The elution of ions is a consequence of sequential grain-scale processes during dry metamorphism (Brimblecombe et al., 1985; Schöndorf and Herrmann, 1987; Goto-Azuma et al., 1994) and also influenced by the pH of meltwater (Goto-Azuma et al., 1994). Moore and Grinsted (2009) calculated the elution factors (e) of several ions present in snow and ice from an ice core drilled at Lomonosovfonna, Svalbard during 1997 (LF-97). According to their results, the most mobile species in the snow were Ca^{2+} , Mg^{2+} , SO_4^{2-} and NO_3^- with Na^+ , Cl^- , NH_4^+ and

A synthetic ice core approach to estimate ion relocation in an ice field site

C. P. Vega et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



A synthetic ice core approach to estimate ion relocation in an ice field site

C. P. Vega et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



K^+ being eluted later. The elution sequence changed slightly in ice, where NO_3^- is the most mobile specie and Mg^{2+} the latest ion to elute. Moore and Grinsted (2009), Ginot et al. (2010) and Eichler et al. (2001) results point to the ionic charge as the control factor in the elution process instead of the acidic character of the ion as reported by Brimblecombe et al. (1985) and Goto-Azuma et al. (1994).

The special characteristics of snow and firn properties on high altitude ice fields with heterogeneous stratigraphy as Lomonosovfonna are a consequence of the long winter season with air temperatures below the freezing point, enhanced snow drift by strong winds, relatively low annual snow accumulation and cold ground surfaces, punctuated by intermittent melt events. The interaction between the meltwater front formed during episodic melt events and the different stratigraphic horizons form preferential flow patterns and create a secondary stratigraphy if refreezing takes place within colder layers (Colbeck, 1991). The secondary stratigraphy produced by refreezing water will have a large influence in meltwater flow and discharge (Bøggild, 2000). During the intermittent melt periods, dislocation of water from more superficial layers percolates into the deeper stratigraphy routed as preferential meltwater flow and the formation of solute enriched ice layers during refreeze of the percolated water will affect the chemistry (i.e. ionic concentrations in this study) recorded in ice cores drilled at the high altitude ice fields of Svalbard.

One estimate to measure if snow melting and percolation have occurred in an ice core stratigraphy is to construct an ion ratio, based on the selection of a pair of ions that originate from the same source but have dissimilar elution coefficients, e.g. Na^+/Mg^{2+} and Cl^-/K^+ as reported by Iizuka et al. (2002) and Grinsted et al. (2006). Using the logarithms of the different ionic ratios as melt indices, denoted as W_{NaMg} or W_{ClK} depending upon the pair of ions selected, a reconstruction of the melting history of a particular ice core site can be reconstructed (Grinsted et al., 2006). Pohjola et al. (2002) found that at Lomonosovfonna, about 25 to 55 % of the annual accumulation over the 20th century suffered melt, with meltwater percolating through the snowpack during warm summers. The most mobile ions reported by Pohjola et al. (2002) were NO_3^- and

SO_4^{2-} , which presented $\sim 50\%$ higher concentrations in ice compared with firn layers. On the other hand, NH_4^+ presented an even distribution between firn and ice. A percolation mechanism was proposed to have an important role in the redistribution of this specie in the ice crystals after deposition. Pohjola et al. (2002) concluded that although some ions may have had a high mobility within the upper part of the LF-97 ice core (0–36 m deep) during anomalously warm summers, the signal was still identifiable, being retained within an annual or biannual resolution.

The aim of this work is to study the temporal change of the snow/firn chemical and physical stratigraphy on the ice field Lomonosovfonna, Svalbard. In this study we will use data from shallow ice cores and snow pits repeatedly studied in 2008, 2009, 2010 and 2011 at Lomonosovfonna to investigate the temporal and vertical change in the stratigraphy and ion composition of the snow/firn column. To manage the sequential change in ion concentrations we will create a *synthetic* ice core using the top layer (~ 1 year accumulation) of different shallow ice cores and snowpits to have a reference record to assess the relocation of major ions by meltwater and the percolation length at the site during recent years. In addition, the melt percentage of the annual accumulation at Lomonosovfonna is obtained by using a positive degree day and a snow-energy model approach, and used together with the *synthetic* core to infer the effects of meltwater on the ionic signals present in recent ice cores drilled at Lomonosovfonna.

2 Methods

2.1 Study sites

Four ice cores (LF-97, LF-08, LF-09 and LF-11) and a 1.50 m deep snowpit (SP LF-10) were considered in this study, all retrieved at the Lomonosovfonna ice cap, which is one of the highest glaciated areas in Svalbard with an elevation of ~ 1250 m a.s.l. (Isaksson et al., 2001; Beaudon, 2012; Vega et al., 2015b) (Fig. 1). Lomonosovfonna is located northeast of Longyearbyen, which is the largest settlement in Svalbard, and

A synthetic ice core approach to estimate ion relocation in an ice field site

C. P. Vega et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Pyramiden, a smaller settlement where coal mining activities were carried out until 1998. Two of the ice cores (LF-97 and LF-08) were drilled at the highest point of the ice cap ca. 100 m apart from each other, while the LF-09 and LF-11 ice cores were drilled ca. 4.5 km east-south-east from the site cored in 1997 and 2008 (Fig. 1, Table 1).

2.2 Sampling and analyses

The cutting, sampling and chemical analyzes of the LF-97 and LF-08 ice cores are reported by Kekonen et al. (2005) and references therein and Beaudon (2012), respectively. The LF-09 and LF-11 ice core cutting and SP LF-10 sampling were done in clean conditions, wearing clean overalls, face masks and powder free gloves. All materials employed to collect the samples were rinsed with ultra-pure water (18 M Ω) and kept in clean plastic bags. Samples and standards were melted and handled under laminar flow hood (class 100) to minimize any contamination from the laboratory environment. Samples and standards were placed in the auto-sampler covered with aluminium foil to avoid any dust contamination. Three sample blanks (made of ultra-pure water) were analysed at the beginning and the end of every batch. Sample checks (bulk-snow from Ny-Ålesund or Uppsala) were analysed every ten samples to ensure the replicability of the measurements within a batch. The analytical error was below 5 % for each ion. Detection limits (D.L.) for each ion were calculated as the average value of six blanks plus 1.68 times the standard deviation (σ) of the six measurements (i.e. $D.L. = \text{average}_{\text{blank}} + 1.68 \times \sigma_{\text{blank}}$), being below $0.3 \mu\text{eq L}^{-1}$ for all ions.

Water stable isotopes ($\delta^{18}\text{O}-\text{H}_2\text{O}$) were analysed at the Institute of Geology at Tallinn Technical University, Estonia. The measurements were done with a Picarro L2120-i water isotope analyser (cavity ringdown spectroscopy technology) with a high precision vaporizer A0211. All isotope measurements were calibrated in a two-point scale against the international standards VSMOW (Vienna Standard Mean Ocean Water) and VSLAP (Vienna *Standard* Light Antarctic Precipitation). The reproducibility of replicate analysis for $\delta^{18}\text{O}-\text{H}_2\text{O}$ measurements was estimated to be $\pm 0.1 \text{‰}$.

A synthetic ice core approach to estimate ion relocation in an ice field site

C. P. Vega et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



past 800 years (Isaksson et al., 2001; Pohjola et al., 2002). An updated time scale for this ice core has been recently published by Divine et al. (2011).

During the 2009 drilling campaign at Lomonosovfonna, two parallel ice cores were drilled: the LF-09 (36 m deep, this study) and a longer core LF-09_{deep} (149.5 m deep) (Wendl et al., 2015). Tritium (^3H) measurements done in the LF-09_{deep} ice core estimate the 1963 ^3H horizon (Pinglot et al., 1999) at 23.6 m w.e., with a resulting accumulation rate of 0.51 m yr^{-1} w.e. between the ^3H radioactive horizon and the top of the ice core (Wendl et al., 2015). In addition, the high resolution chemical data available for the LF-09 ice core (samples taken each 8 cm) allowed the use of a multilinear regression method (MLR) developed by Moore et al. (2012) to account for volcanic layers in the non-sea salt sulfate concentrations. A detailed description on the usage of the MLR method on the dating of the LF-09 ice core can be found in Vega et al. (2015b). In addition to the MLR method, an automated $\delta^{18}\text{O}-\text{H}_2\text{O}$ cycles counting routine was used in the dating. This method counted $\delta^{18}\text{O}-\text{H}_2\text{O}$ annual cycles that had an amplitude $A > 0.1 \text{ ‰}$ ($\delta^{18}\text{O}$ uncertainty in the SMOW scale) and a frequency in the sub-annual cycle (λ_{seas}) larger than 1/3 the accumulation rate (Pohjola et al., 2002). Using both methods, the time scale for the LF-09 core was estimated to span between 1957–2009 (Vega et al., 2015b).

The LF-08 and LF-11 ice cores were dated using the automated $\delta^{18}\text{O}-\text{H}_2\text{O}$ annual cycles counting routine and by comparing the Cl^- record with the respective LF-09 ice core profile. The time scales for the cores were estimated to cover the 1989–2008 and 2004–2011 periods for the LF-08 and LF-11 (Vega et al., 2015a) ice cores, respectively. The time scale of the SP LF-10 snowpit was obtained by assuming a constant snow accumulation rate over the depth-scale, resulting in a time coverage between 2009.4–2010.4.

TCD

9, 5063–5095, 2015

A synthetic ice core approach to estimate ion relocation in an ice field site

C. P. Vega et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



3.2 Major ions

All the ionic concentrations were binned to annual averages to obtain equally spaced time series of one year resolution, which can be compared in between the different ice cores. Having all the chemical species as annual averages, their concentrations (c_i) were normalized to the mean value for each specie in each ice core record, during the total overlapping period (1957–2009) to compare the variations of the ions in the different ice cores (Eq. 1).

$$c_N = \frac{c_i - c_{\text{mean}}}{\sigma}, \quad (1)$$

where c_N is the normalized concentration of a given ion, c_{mean} is the ion mean in the specific ice core or strata, and σ is the standard deviation of the series.

When comparing the annual average concentrations between the different ice cores, some of the peaks show a lag between the different records which likely is associated to the uncertainty on the dating of the three different ice cores. We applied a 5 year moving average smoothing to the annual chemical data (Fig. 2). The dating error of the LF-08 and LF-09, in respect to the LF-97 ice core, was estimated as ± 2 years. We found that similar temporal variations experienced by the different ions were registered in the different Lomonosovfonna ice cores during the overlapping period studied (Fig. 2).

Table 3 shows the Pearson R values between the different species in the LF-97 and LF-09 ice cores and the LF-09 and LF-09 ice cores, smoothed out with 5 year moving averages. The R values between the LF-97 and LF-08 ice cores were not calculated since the overlapping period using 5 year moving average was too short. The R values in Table 3 suggest that the LF-97 and LF-09 ice cores have comparable records for all ions. However, the correlation coefficients measured in the LF-08 and LF-09 ice cores are not significant at the 95 % confidence interval (with the exception of Mg^{2+} , $R = -0.53$).

A synthetic ice core approach to estimate ion relocation in an ice field site

C. P. Vega et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



This can be interpreted as SO_4^{2-} eluting steadily and further down the snowpack than the other ions during this period (Fig. 8).

We estimate a *minimum* ionic relocation length as the difference in depth between the maximum/minimum (deposition/elution) c_N peaks, as shown in Fig. 8 and presented in Table 4. The relocation length was calculated for all ions, with the exception of SO_4^{2-} which does not show an elution peak that is significant within the time window studied. For the ions that show more than one significant deposition/elution peak, the relocation length calculated considering the second maximum/minimum c_N peak is also shown in Table 4.

Considering the deposition/elution c_N peaks estimated above, the ionic elution scheme at Lomonosovfonna for the last 5 years results in SO_4^{2-} being the most mobile ion and NO_3^- the least mobile one. This elution scheme differs with previous studies done at Lomonosovfonna (Moore and Grinsted, 2009) mainly in locating NO_3^- at the end of the elution sequence. However, it should be noticed that we estimated two possible relocation lengths based in the depth difference between deposition/elution peaks (as explained before). Therefore, considering a NO_3^- relocation length of 1.16 m, the elution sequence agrees with previous findings by Moore and Grinsted (2009), locating the acidic ions (SO_4^{2-} and NO_3^-) as more mobile than the others ions. The elution sequence and ion relocation lengths shown in Table 4, suggest that the less mobile ions are relocated by the meltwater at short lengths, probably trapped in thin and medium-thick ice layers present within the ice column observable in the LF-97, LF-09 and LF-11 cores (Fig. 9), or in refrozen water that soaks the firn column filling the pore space. More mobile ions may percolate further down in the snowpack and get trapped in medium-thick to thick ice layer which start to form at about 3–10 m deep in the ice column as observable in the LF-97 and LF-09 cores (Fig. 9).

In order to estimate how the ionic relocation length during 2007–2010 is connected to meltwater percolation length and snowpack melting, we have estimated the melt percentage at Lomonosovfonna using two different methods, as shown in the following section.

A synthetic ice core approach to estimate ion relocation in an ice field site

C. P. Vega et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



A synthetic ice core approach to estimate ion relocation in an ice field site

C. P. Vega et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



(Table 5) were calculated using the results in Figs. 10 and 11 and the annual accumulation rate between 2007–2010 at Lomonosovfonna obtained from the LF-09 and LF-11 cores. The melt percentage (MP) obtained by using the instrumental temperatures (48 % for the period 2007–2010) was in the order of previously reported values of 47 % of melt during the warmest years and 28 % during the coldest, during the period 1976–1995 (Pohjola et al., 2002). The relatively high MP values obtained in this study for the period 2007–2010 can be explained by the increase in temperatures at the site with a consequent steady increase of the number of PDD at the summit of Lomonosovfonna at a rate of 0.4 day yr^{-1} for the period 1979–2012 in which instrumental temperatures exist in all the three stations used, Sveagruva, Ny-Ålesund and Longyearbyen airport; the MPs calculated using the PDD approach over the period 1979–2010 show a steady increase of $1 \% \text{ yr}^{-1}$ in line with the trend in decreasing the annual snow accumulation in -0.01 m yr^{-1} w.e. during the same period (accumulation data from the LF-09 ice core).

However, the calculated MPs using the PDD approach and the snow-energy do not agree with the results obtained by the synthetic core approach which suggest moderate melting during the 2007–2010 period which is also supported by the stratigraphy observed in the LF-09 and LF-11 cores (Fig. 9).

Due to the dissimilar results of MP obtained using the PDD and snow-energy model approaches (Table 5), we compared the density profiles of the shallow ice cores used in this study (LF-08, LF-09 and LF-11) with the total layer density values obtained using a simple firn-densification model (Reeh et al., 2005). The ice core density profiles were binned in annual averages in order to be comparable with the output of the firn-densification model. As described by Reeh et al. (2005), the model requires both annual average temperatures and snow accumulation at the study site as input, which were set as -18.3°C and 0.39 m w.e. , according to instrumental temperatures from Sveagruva, Svalbard Airport and Ny-Ålesund (the lapse rate employed to calculate the temperatures at the Lomonosovfonna summit was $-0.0044^\circ\text{C m}^{-1}$), and average accumulation rates obtained from the LF-09 and LF-11 ice cores, respectively, for the pe-

index during the ice core time span. Therefore, the firn-densification model does not include the variability in meltwater production at Lomonosovfonna through time which is key to the understanding of ion relocation within the snowpack at long time scales; however, the MP estimated here are suitable to understand the ionic relocation and water percolation during the period 2007–2010.

Figure 13 shows a comparison between the LF-11 and the LF-syn ice core depth-time scale. It is clear from Fig. 13 that the depth differences between the ice cores (black and grey lines) can be related to a partial melting of the snowpack and refreezing of the meltwater taking place between the 2007–2011 period, as evidenced by the ionic relocation. An estimate of the decrease in depth of the LF-syn ice core by the effects of snowpack melting considering a MP = 20, 30, 48 and 70 % during 2007–2010 as suggested by the firn-densification model, MPs obtained using the PDD approach (using instrumental temperatures) and the snow-energy model is shown in Fig. 13. It can be observed that a MP = 20–30 % results in a LF-syn ice core depth-time scale highly similar to the depth-time scale of the LF-11 ice core during the 2007–2010 period, confirming that the MP during that period is coherent with the results of the firn-densification model but not with the PDD results shown in Table 5. This relatively low MP reinforces the fact that during this period, ion relocation took place at moderate depths (0.5–2 m) and that the meltwater percolation depth was most probably in the same order, with refreezing of meltwater within the snowpack pore space or by forming thin ice layers (0.03–0.15 m) (Fig. 9).

4 Conclusions

By comparing different ice cores from Lomonosovfonna and using a *synthetic* ice core approach, we have been able to estimate the elution signal and relocation length of different ions measured at the study site, in order to assess the transfer function between the atmospheric ionic concentrations and the concentration in snow and ice.

A synthetic ice core approach to estimate ion relocation in an ice field site

C. P. Vega et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



percentages inferred for the 2007–2010 period compared with the whole period covered by the LF-09 core are most probably a result of a combination of relatively high snow accumulation rates and reduced PDD during the 2007–2010 period despite of the warmer conditions registered during the last decades.

5 Considering our findings, we conclude that despite of the warmer conditions and higher number of PDD registered at Lomonosovfonna during the last decades, the ionic signal affected by melting is retained within the same year of deposition for all major ions, with the exception of SO_4^{2-} which can possibly be re-deposited in previous annual layers.

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A synthetic ice core approach to estimate ion relocation in an ice field site

C. P. Vega et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



A synthetic ice core approach to estimate ion relocation in an ice field site

C. P. Vega et al.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

Claremar, B., Obleitner, F., Reijmer, C., Pohjola, V., Waxegård, A., Karner, F., and Rutgersson, A.: Applying a mesoscale atmospheric model to Svalbard Glaciers, *Adv. Meteorol.*, 2012, 321649, doi:10.1155/2012/321649, 2012.

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A synthetic ice core approach to estimate ion relocation in an ice field site

C. P. Vega et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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A synthetic ice core approach to estimate ion relocation in an ice field site

C. P. Vega et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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A synthetic ice core approach to estimate ion relocation in an ice field site

C. P. Vega et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Table 3. Pearson coefficients (R) at a 95% confidence interval, between the different ions measured in the LF-97 and LF-09 ice cores (normalized concentrations and smoothed out as 5 year running averages). Significant values ($p < 0.05$) are shown in italics. The overlapping period is 1957–1997.

LF-97 and LF-09 ice cores								
Ion	Cl ⁻	NO ₃ ⁻	SO ₄ ²⁻	Na ⁺	NH ₄ ⁺	K ⁺	Ca ²⁺	Mg ²⁺
R	0.46	0.64	0.56	0.49	0.77	0.66	0.36	0.72
LF-08 and LF-09 ice cores								
R	-0.21	0.02	0	-0.30	0.32	-0.16	-0.23	-0.53

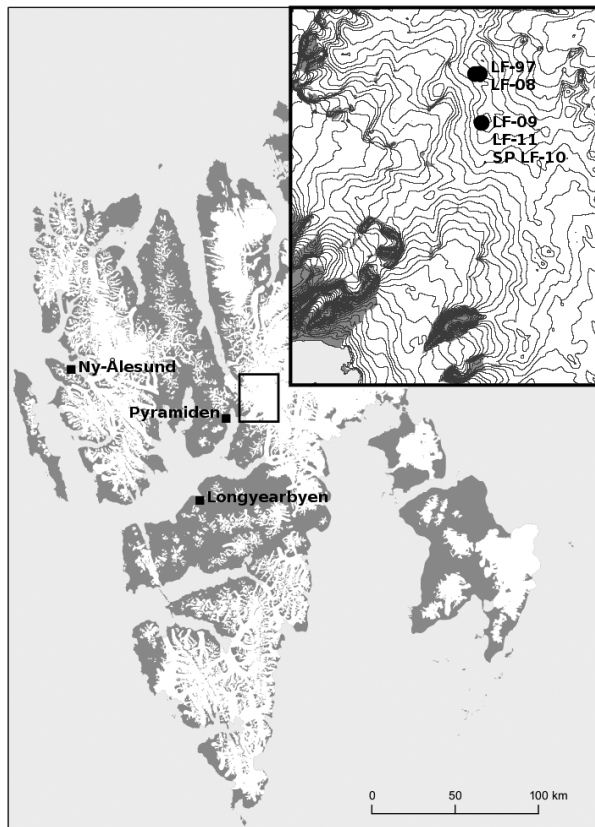


Figure 1. Map of Svalbard showing the main settlements: Longyearbyen, Ny-Ålesund and Pyramiden, and the LF-97, LF-08, LF-09 and LF-11 drilling sites at Lomonosovfonna.

A synthetic ice core approach to estimate ion relocation in an ice field site

C. P. Vega et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



A synthetic ice core approach to estimate ion relocation in an ice field site

C. P. Vega et al.

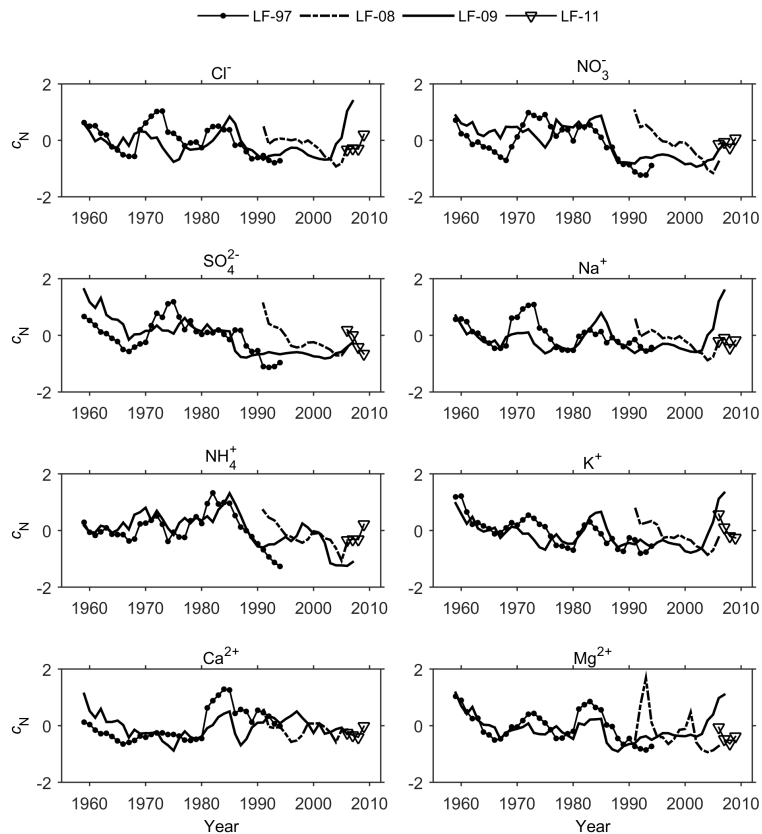


Figure 2. Comparison of normalized ionic concentrations (c_N) in the different LF-ice cores using a 5 year moving average.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



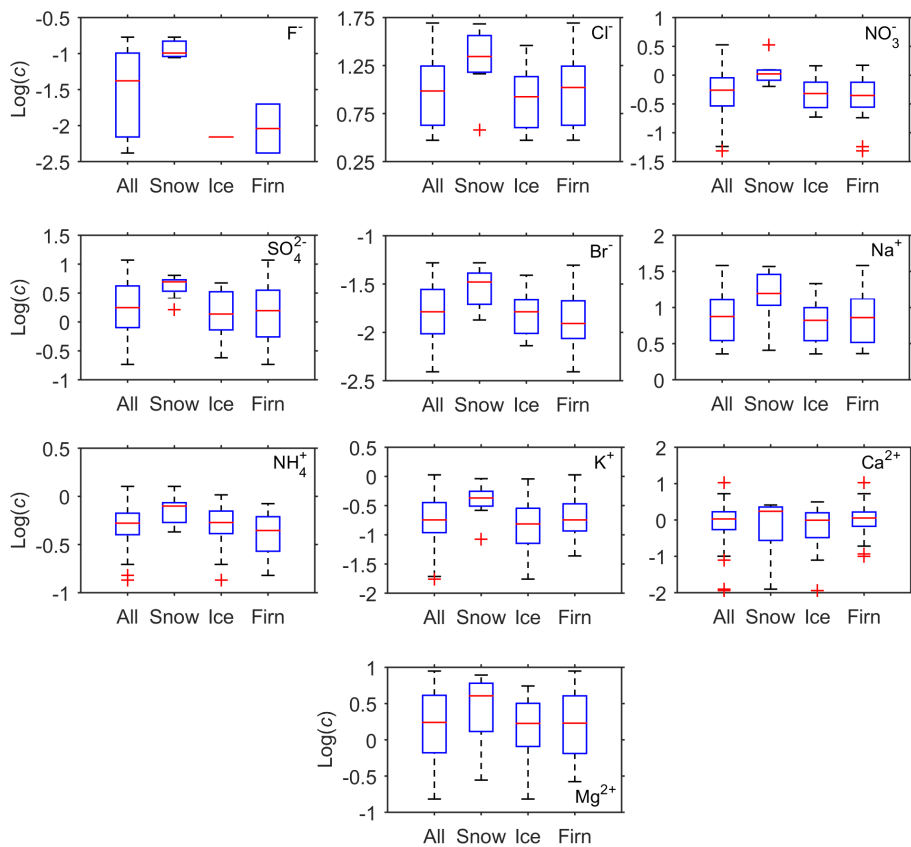


Figure 3. Box plot of log-transformed ionic concentrations ($\log(c)$) found in the different stratigraphic units in the LF-09 ice core between 2004 and 2009.

A synthetic ice core approach to estimate ion relocation in an ice field site

C. P. Vega et al.

Title Page

Abstract Introduction

Conclusions References

Tables Figures

◀ ▶

◀ ▶

Back Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



A synthetic ice core approach to estimate ion relocation in an ice field site

C. P. Vega et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

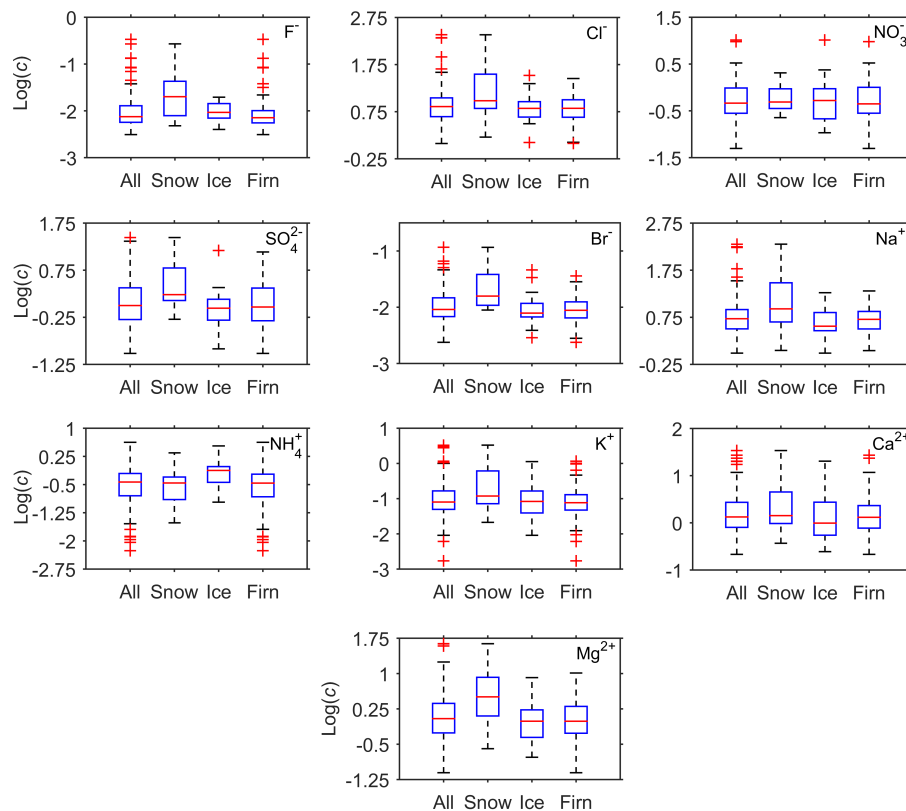


Figure 4. Box plot of ionic concentrations ($\log(c)$) found in the different stratigraphic units in the LF-11 ice core between 2004 and 2011.

A synthetic ice core approach to estimate ion relocation in an ice field site

C. P. Vega et al.

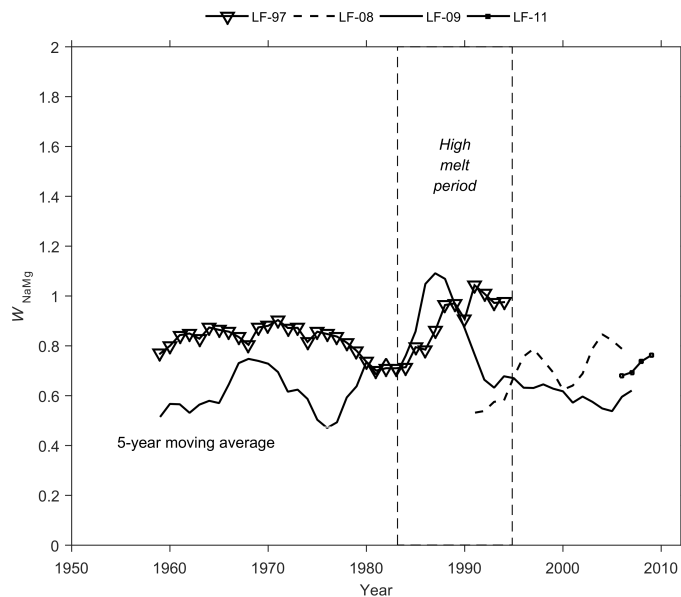


Figure 5. Melt index W_{NaMg} calculated in the different Lomonosovfonna ice cores. The values correspond to 5 year moving averages. The dashed rectangle indicates a high melting period during mid-80s to mid-90s.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



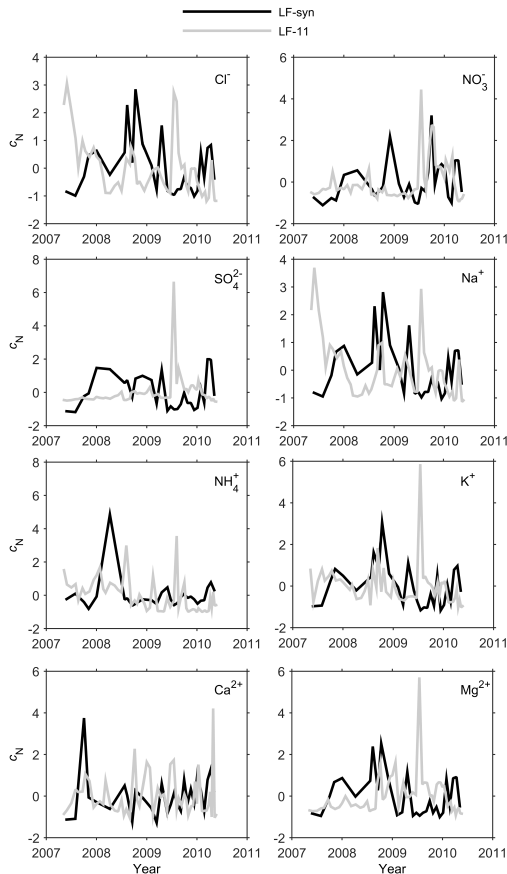


Figure 7. Comparison between a *synthetic* ice core (LF-syn, black) and the LF-11 ice core (grey). Ionic concentrations are normalized (c_N) to mean values according to the LF-syn ice core time scale.

A synthetic ice core approach to estimate ion relocation in an ice field site

C. P. Vega et al.

Title Page

Abstract Introduction

Conclusions References

Tables Figures

◀ ▶

◀ ▶

Back Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



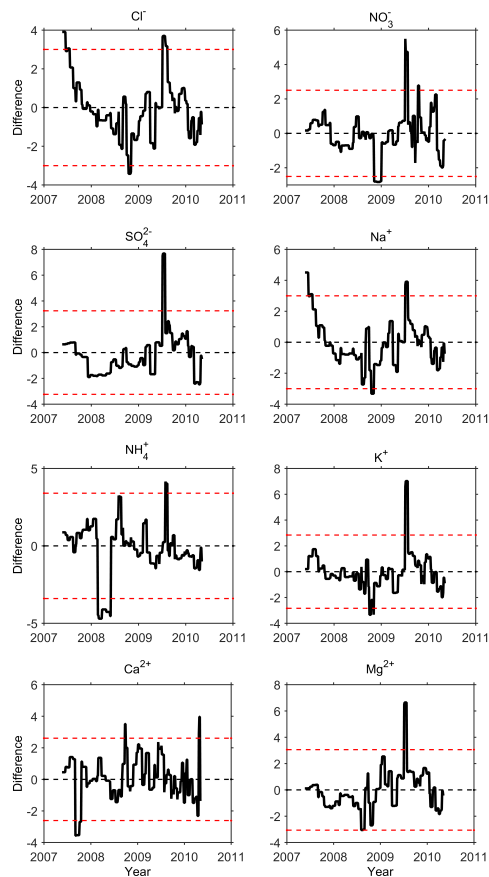


Figure 8. Difference between the normalized ionic concentrations (c_N) in the LF-11 and the LF-syn ice cores (black line). Twice the standard deviation (dashed red line) and zero values (dashed black line) are also shown. Deposition is a positive excursion and elution zones are negative excursions from the mean in each diagram.

A synthetic ice core approach to estimate ion relocation in an ice field site

C. P. Vega et al.

Title Page	
Abstract	Introduction
Conclusions	References
Tables	Figures
◀	▶
◀	▶
Back	Close
Full Screen / Esc	
Printer-friendly Version	
Interactive Discussion	



A synthetic ice core approach to estimate ion relocation in an ice field site

C. P. Vega et al.

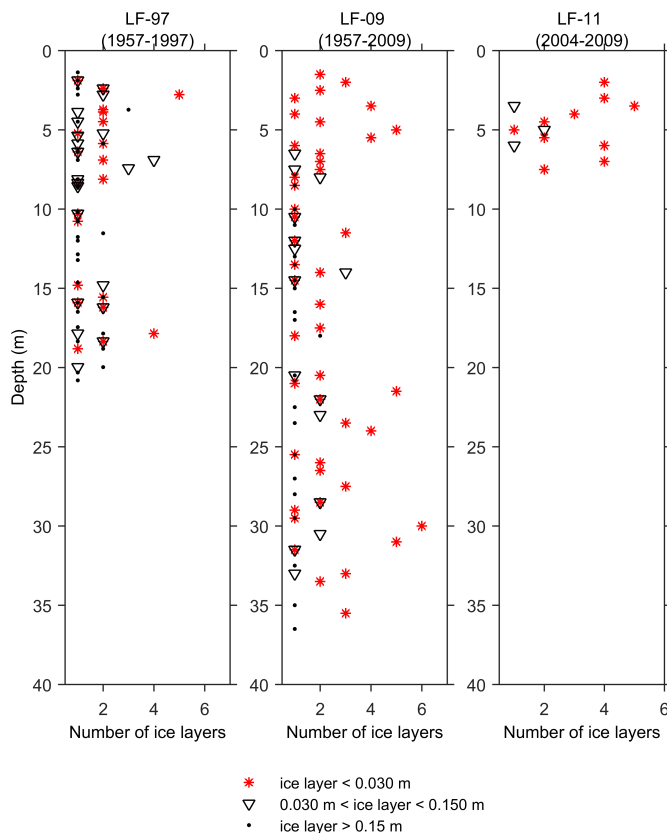


Figure 9. Number of ice layers found at different depths in the: LF-97 (left panel), LF-09 (centre panel) and LF-11 (right panel) ice cores. The ice layers were divided in three classes according to their thickness: thin layers (*), medium-thick layers (▽) and thick layers (●).

[Title Page](#)
[Abstract](#)
[Introduction](#)
[Conclusions](#)
[References](#)
[Tables](#)
[Figures](#)
[Back](#)
[Close](#)
[Full Screen / Esc](#)
[Printer-friendly Version](#)
[Interactive Discussion](#)

A synthetic ice core approach to estimate ion relocation in an ice field site

C. P. Vega et al.

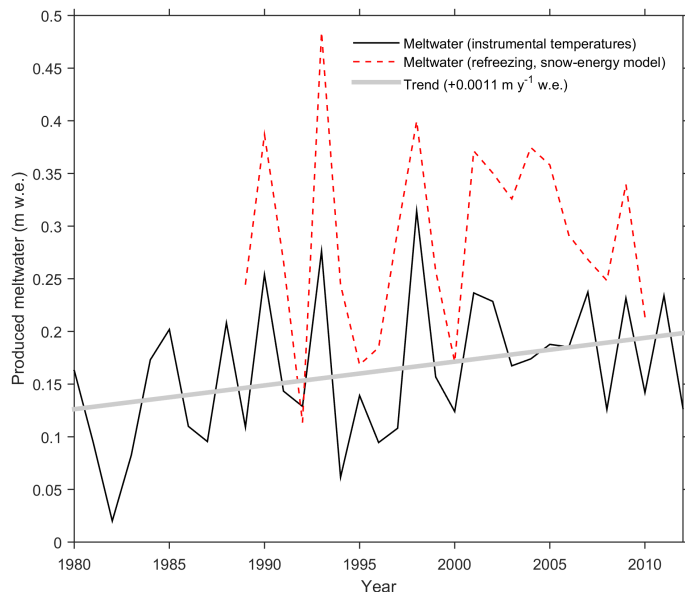


Figure 10. Produced meltwater calculated at Lomonosovfonna using the PDD (black) and snow-energy model (dashed red) approaches. The linear trend of meltwater calculated with the PDD approach and instrumental temperatures is also shown over the 1979–2012 period (grey).

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

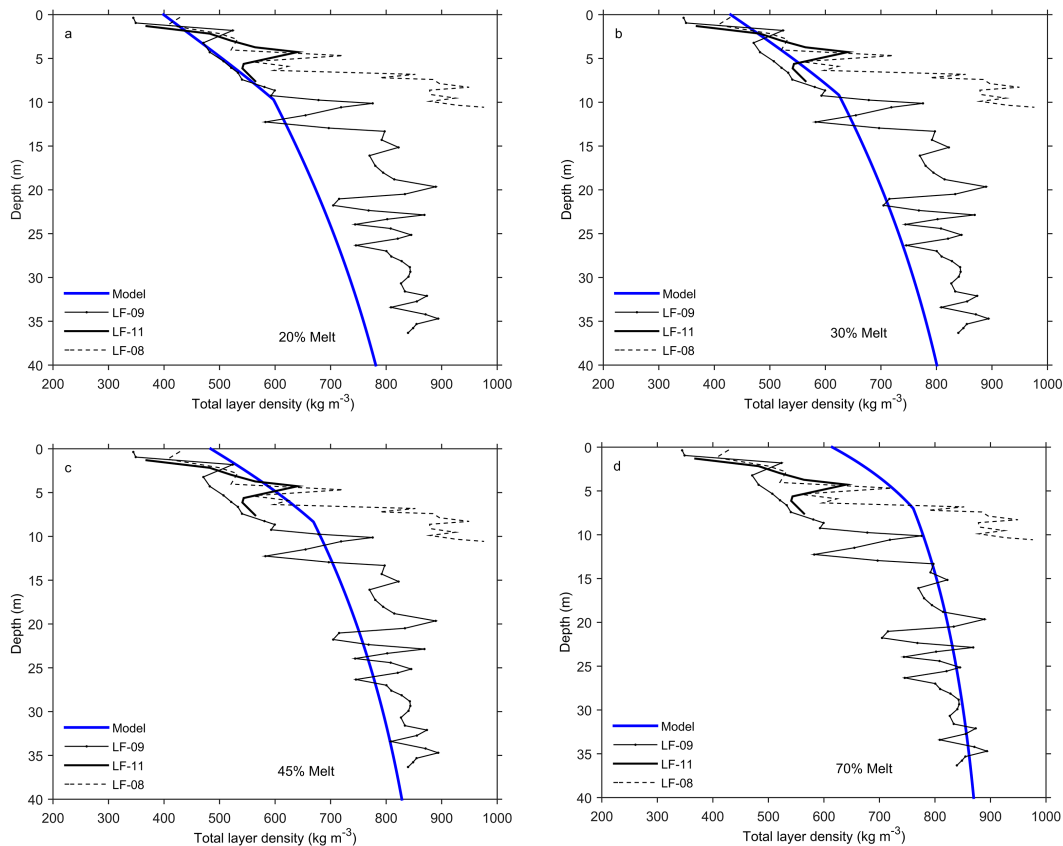


Figure 12. Comparison between the LF-08, LF-09 and LF-11 ice core density profiles (annual averages) and the output of the firn-densification model (Reeh et al. 2005), considering: **(a)** 20 % of melt, **(b)** 30 % of melt, **(c)** 45 % of melt, and **(d)** 70 % of melt.

A synthetic ice core approach to estimate ion relocation in an ice field site

C. P. Vega et al.

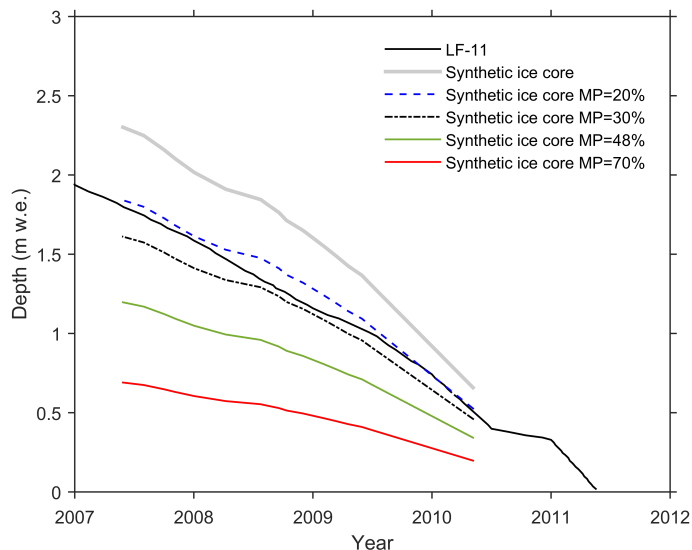


Figure 13. Depth–time scales for the LF-11 and LF-syn ice cores during 2007–2011 (black and grey lines, respectively). Blue dashed, black-dot-dashed, green and red lines represent the synthetic core depth–time scale considering a MP of 20, 30, 48 and 70 %, respectively.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)