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Variability, origin and physical characteristics of dust aerosol in Iceland

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Variability, origin and physical characteristics of dust aerosol in Iceland

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Dissertation submitted in partial fulfillment of a
Philosophiae Doctor degree in Physical Sciences

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Abstract

Emissions of particulate matter from the natural sources account for a significant part of the total particulate air pollution. Less industrialized and inhabited regions, such as deserts, can contribute remarkably to air pollution. Iceland is a good example of such areas. The trends in long-term dust activity in Iceland and results from the pioneer field experiments on dust atmospheric and snow measurements are presented in this thesis. Long-term dust frequency in Iceland is based on the meteorological data from 30 weather stations in period 1949-2011. Dust day frequency in Iceland is similar to the major desert areas of the world (Mongolia, Iran, USA, China). Frequent volcanic eruptions with the re-suspension of volcanic materials and dust haze increase the number of dust events fourfold, resulting in 135 dust days annually. The Sea Level Pressure oscillation controlled whether dust events occurred in NE or in southern part of Iceland. The Arctic dust events (NE Iceland) were typically warm and during summer/autumn (May-September) while the Sub-Arctic dust events (S Iceland) were mainly cold and during winter/spring (March-May). A total of 32 severe dust storms (visibility < 500 m) was observed. Dust deposition of 31 – 40 million tons influences areas of > 500,000 km², while some dust plumes are spanning > 1000 km at times. These results confirm that Icelandic dust sources are the most active in the Arctic/sub-Arctic region. Dust is also distributed over glaciers (about 4.5 million t annually) and oceans (6 – 14 million t annually). Our measurements show that Icelandic dust deposited on snow lowers the snow albedo and reduces the snow density as much as Black Carbon, the most powerful absorbing aerosol. The dust has a marked influence on Icelandic ecosystems. The oceanic deposition of the iron-rich dust can potentially affect the primary productivity in oceans around Iceland, especially in spring and late summer. The investigations of physical properties of volcanic dust reveal major differences in mineralogy, geochemical compositions, shapes, sizes and colour, compared to the crustal mineral dust. Icelandic dust is of volcanic origin, dark in colour with sharp-tipped shards and large bubbles. About 80 % of the particulate matter is volcanic glass rich in heavy metals, such iron and titanium. Suspended dust measured at the glacial dust source consists of extreme numbers of close-to-ultrafine particles with a similar numbers as reported during the active eruptions. However, giant Icelandic particles (50 – 100 µm), can travel long distances. Suspended grains > 2 mm were captured during the severe dust storm after the Eyjafjallajökull eruption in 2010, when the aeolian transport exceeded 11 t of materials over one meter wide transect. This places Icelandic dust storms among the most extreme wind erosion events recorded on Earth. First experiments on optical properties of volcanic dust in the laboratory show that Icelandic dust has same or lower spectral reflectance than Black Carbon. This indicates that climate forcing of Icelandic volcanic dust is different to that concluded for mineral dust in climate change predictions. Icelandic volcanic dust tends to act as a positive climate forcing agent, both directly and indirectly. The high frequency, severity and year-round activity of volcanic dust emissions suggest that Icelandic dust contributes to Arctic warming. Iceland is not only a substantial source for local air pollution; it has likely long-term effects on the Arctic and European air pollution.

Útdráttur

Stór hluti rykframleiðslu veraldar kemur frá náttúrulegum ryk uppsprettum. Mörg strjálbýl svæði jarðar og eyðimerkur gefa frá sér mikið ryk. Ísland er á meðal slíkra svæða. Í þessari ritgerð er greint frá þróun rykmengunar í Íslandi út frá rannsókn á langtímagögnum frá veðurmælingum og gerð er grein fyrir tilraunum til að mæla rykmengun í andrúmslofti og áhrif ryks á snjó. Birt eru fyrstu gögn um tíðni og virkni rykstorma á Íslandi sem og upplýsingar um smásjáreiginleika rykefna, fjölda og massa örefna (particular matter). Einnig er rykframleiðsla frá Íslandi ákörðuð magnbundið, sem og áfok á land, sjó og jökla. Gefin eru dæmi um mjög aflmikla vindrofsatburði. Langtímamælingar eru byggðar á veðurmælingum á veðurstöðvum 1949-2011. Tíðni fokatburða svipar til tíðni þeirra á helstu eyðimerkursvæðum jarðar, t.d. Mongólíu, Íran, USA og Kína. Eldgos og gerð fokefnanna eykur tíðnina fjórfalt og leiðir til 135 rykdaga á landinu á ári. Rykframleiðslan nemur 31-40 milljónum tonna sem falla á >500 000 km² hvert ár, en sumir strókarnir ná >1000 km burt frá landinu. Niðurstöðurnar sýna að landið er virkasta ryksvæðið á arktískum svæðum jarðar. Um 4,5 milljón falla jökla en 6-14 milljón t á sjó við landið. Rannsóknirnar sýna að rykið, sem er dökkleitt, minnkar endurgeislun af snjó meira en sót (black carbon), sem annars hefur talið mikivirkasta efnið við að draga í sig sólarljós. Rykið er járnríkt og kann að hafa mikil áhrif á frumframleiðni í sjó og lífríki hans. Rannsóknir á eðliseiginleikum ryksins sýna að það er afar frábrugðið ryki frá meginlöndunum er varðar efnasamsetningu, lögun, stærð og lit. Um 80% ryksins er dökkt basaltgler sem inniheldur mikið af málum á borð við járn og títaníum. Ryk frá jökuljöðrum inniheldur mikið af mjög fínum efnum sem svipar til rykmengunar frá eldgosum. En þó bersta einnig afar stór korn (50-100 micrometrar) langar leiðir. Gjóskukorn stærri en 2 mm berast með vindi en vindrof flutti > 11 t efnis yfir eins m breiða línu í miklum stormi á Skógaheiði eftir gosið. Þetta er trúlega einn mesti sandstormur sem hefur verið mældur á jörðinni. Íslenskt ryk hefur svipaða endurkastseiginleika og sót, sem kann að gera rykið að jákvæð loftslagsbreyta, öfugt við annað ryk. Líklegt er að ryk frá Íslandi auki á hlýnum arktískra svæða vegna mikillar tíðni og virkni rykstorma á Íslandi. Rykið veldur ekki aðeins loftmengun á Íslandi heldur hefur líklega áhrif á loftæði á arktískum svæðum og í Evrópu.

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Abbreviations

| | |
|----------|---|
| ADMI | Atmospheric Dust Measurements in Iceland campaign |
| ASD | Analytical Spectral Device Spectrophotometer |
| AUI | Agricultural University of Iceland |
| BC | Black Carbon |
| CNES | Centre National d'Etudes Spatiales, France |
| COST | European Cooperation in Science and Technology |
| CIMAS | Cooperative Institute for Marine and Atmospheric Studies |
| CRAICC | Nordic Centre of Excellence for Cryosphere-Atmosphere Interactions in a Changing Arctic Climate |
| DD | Dust day |
| EDX SEM | Energy Dispersive Spectrometry |
| EIA | Environmental Agency of Iceland |
| ECMWF | European Centre for Medium-Range Weather Forecasts |
| EGU | European Geoscience Union |
| FIGIFIGO | Finnish Geodetic Institute Field Goniospectrometer |
| GISP2 | Greenland Ice Sheet Project |
| HYSPLIT | Hybrid Single Particle Lagrangian Integrated Trajectory Model |
| IMO | Icelandic Meteorological Office |
| IPCC | Intergovernmental Panel on Climate Change |
| LOAC | Light Optical Aerosols Counter |
| MODIS | Moderate Resolution Imaging Spectroradiometer |
| NAMEE | Northern Africa-Middle East-Europe Regional Center |
| NILU | Norwegian Institute for Air Research |
| OPS | Optical Particle Sizer |
| PM | Particulate Matter |
| PMF | Positive Matrix Factorization |
| RMD | Reference Material Database |
| SENSIT | Wind Eroding Mass Sensor |
| SOS | Soot on Snow campaign |
| UST | Environmental Agency of Iceland |
| UV | Ultraviolet light |
| WHO | World Health Organization |
| XRD | X-ray Powder Diffraction |

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1 Introduction

There is a growing concern about the influence of dust on Earth's ecosystems and humans and the research reported here focuses on dust production from Icelandic deserts. This dissertation has relevance to a wide range of topics, such as air quality, aeolian processes, atmosphere-cryosphere interactions, meteorology and long-range transport of aerosol, climate, ecosystem development (both ocean and land). The main emphasis is given to atmospheric aerosols, mainly dust, in a non-polluted Arctic/sub-Arctic region, Iceland. This study deals with the dust event frequency and severity, dust suspension processes at a dust source, dust deposition over oceans, glaciers, on snow, and physical characteristics of the aerosol.

1.1 Background

Iceland is considered as a country with generally clean and less polluted air than more densely populated countries in Europe and North America. Large areas of the land are, however, volcanic sandy deserts and ice-proximal areas, producing high amounts of natural air pollution (Arnalds, 2010). The areas with limited vegetation cover are >22 % of Iceland and >40% of the country is classified with considerable to very severe erosion (Arnalds et al., 2001). Emissions from these sources, enhanced by strong winds, affect regional air quality in Iceland, such as in the capital Reykjavik (Thorsteinsson et al., 2011). Particulate Matter (PM₁₀- for particles with diameter <10 µm) concentrations during dust events in Reykjavik often exceed the health limit of 50 µg m⁻³ over 24 h (WHO, 2005; UST, 2014a). PM concentrations measured during dust events in vicinity of dust sources (<30 km) exceed the health limit in order of 10-100 times (UST, 2014a). Such high concentrations are likely to affect human health. Research has showed that mortality increases about 8 % per 10 µg m⁻³ when Saharan dust is suspended (Perez et al., 2008). Thus, natural air pollution has negative impacts on health of humans or livestock, and needs to be determined not only for highly populated areas, but also for less densely populated and isolated areas such as Iceland.

Natural dust is emitted from many desert areas on Earth. The highest frequency of dust events is in Africa and Australia where the major deserts are located (N'TchayiMbourou et al., 1997; Ekström et al., 2004). Asian deserts such Gobi and Iranian deserts have lower dust frequency than Sahara desert (Qian et al., 2002; Natsagdorj et al., 2003; Jamalizadeh et al., 2008). The lowest frequency is reported from the central deserts in the USA (Steenburgh et al., 2012). South American deserts are also among active dust sources (Lamy et al., 2014).

Although dust is most often associated with dry and warm desert areas, dust is frequently emitted in cold climate regions and at high latitudes such Iceland, Alaska, Greenland, Svalbard and Antarctica (Nickling, 1978; Arnalds, 2010; Dornbrack et al., 2010; Lancaster et al., 2010; Crusius et al., 2011; Prospero et al., 2012; Bullard, 2013). Polar deserts with

glacially-derived sediments of riverbeds or ice-proximal areas contribute to the dust cycle (Bullard, 2013).

Iceland is an important source of volcanic sediments that are subjected to intense aeolian activity (Arnalds, 2010; Prospero et al., 2012; Thorarinsdottir and Arnalds, 2012) and is likely the largest glaciogenic dust source area in the Arctic/sub-Arctic region. Dust deposition rates are exceeding $500 \text{ g m}^{-2} \text{ yr}^{-1}$ in some areas (Arnalds et al., 2010). Seven major dust sources have been identified (Arnalds, 2010). These sources are all in vicinity of glaciers. The most active glacial flood plain, Dyngjúsandur, covers an area of about 270 km^2 with up to 10 m thick sediments (Mountney and Russell, 2004) and is the main source for dust events in Northeast Iceland and towards Arctic. The major dust sources in South Iceland are Skeidararsandur, Myrdalssandur, Mælifellssandur, Landeyjasandur, resulting in dust events south towards Europe during northerly winds, but alternatively towards Reykjavik and in direction of North America during easterly winds. The Hagavatn plume area is the source for dust events towards Reykjavik and the ocean southwest of Iceland. Volcanic eruptions are frequent in Iceland, with individual volcanic events occurring on average at a 3-4 year interval, with frequent production of high amounts of tephra (Gudmundsson et al., 2008). Deposited volcanic ash is subjected to intense aeolian processes and often resuspended (Arnalds, 2010, 2013; Leadbetter et al. 2012; Thorsteinsson et al., 2012; Bullard, 2013). Volcanic inputs are also important contributors to dust events in Iceland.

Global dust differs in mineralogical and geochemical composition. The major deserts such as Sahara consist of crustal dust which is of quartz-rich materials (Marconi et al., 2014). Icelandic dust differs from dust originating in continental dust sources. It is volcanogenic in origin, of basaltic composition, with lower SiO_2 proportions ($<50\%$) and higher Al_2O_3 , Fe_2O_3 , CaO contents than crustal dust (Baratoux et al., 2011). Sediments from glacial plains and sandy deserts are primarily poorly crystallized basaltic materials (glass) containing high quantities of iron, which can have a substantial impact on the ocean chemistry and fertility (Arnalds, 2010). However, mineralogical and geochemical compositions of dust from seven different dust sources in Iceland are not identical (Baratoux et al., 2011); similarly the composition of tephra differs between Icelandic volcanoes (Óladóttir et al., 2011). Icelandic volcanic dust made of glass can be extremely sharp and contain large bubbles, allowing particles as large as $50 \text{ }\mu\text{m}$ to travel long distances (Navratil et al. 2013).

Satellite images have shown that dust particles in dust plumes are transported over the Atlantic Ocean and Arctic Ocean $> 1000 \text{ km}$ at times (Arnalds, 2010). Globally, fine dust particles may be transported at altitudes of up to 10 km and can be carried distances of $>10,000 \text{ km}$ (Husar, 2004). Grousset et al. (2003) suggested that dust particles can travel over a $20,000 \text{ km}$ in two weeks. Icelandic dust is likely to contribute to Arctic or European air pollution and affect indirectly the climate via dust deposition on Arctic glaciers or sea ice. Local glaciers cover about 11% of Iceland, but the closest distance of Greenland's glacier is about 500 km from Iceland. Studies of dust deposition on snow show that dust mixed in snow accelerates snowmelt through its direct reduction of snow albedo and further indirect reduction of albedo by accelerating the growth of snow grain size (Painter et al., 2012). They showed that mean spring dust radiative forcing ranged between 45 and 75 W m^{-2} , which reduced snow cover duration by 21 to 51 days. However, direct radiative

forcing of mineral dust is calculated as negative in the IPCC report (IPCC, 2013). It is therefore important to investigate indirect forcing of dust deposited on snow in a greater detail. Volcanic dust from Iceland is dark in contrast to the crustal silica, which is light in colour, and might therefore have opposite radiative characteristics to crustal dust. The location of Iceland in Arctic/sub-Arctic region may therefore enhance the climate effects of dust.

Dust storm events can have a direct negative impact on ecosystems by burial of vegetation, and by causing loss of fertile topsoil (Fields et al., 2010; Arnalds, 2013). Icelandic authorities have undertaken several land reclamation measures since the 1940s to reduce the negative effects of dust activity and production (Crofts, 2011). Iceland is an island and dust deposition occurs also over the surrounding seas (Arnalds, 2010). Achtenberg et al. (2013) measured significantly elevated iron levels in the sea south of Iceland during the 2010 Eyjafjallajökull eruption. This suggests that Icelandic dust events could have positive impacts on the North Atlantic and Arctic Ocean via natural iron fertilization.

1.2 Motivation and objectives

The Environmental Agency of Iceland (EAI) reports the outdoor air quality in Iceland as “generally quite good” due to an oceanic climate and steady winds (UST, 2014b). The focus on an impaired visibility due to natural emissions in many places of Iceland led to the research and this thesis. Winds in Iceland are frequently of high velocities leading to suspension of large dust quantities that the regional haze changes into a dust storm event. The main objectives of the research were to quantify these dust events and understand the nature and impacts of dust production in Iceland.

Long-term measurements of dust aerosol in Iceland are not available. There is a small network of the PM stations (seven stations of which four are in Reykjavik, in operation mostly from 2010) around Iceland maintained by the EAI. Large areas, especially in vicinity of dust sources, have never been measured in terms of the PM air pollution. The measurements provide only the $PM_{(1-10)}$ mass concentrations, missing the information on the particle number concentrations. The PM masses are measured far from the source of air pollution and only on ground. The second goal of the present work is to provide such measurements to understand the physical characteristics of fine dust aerosol. In addition, chemistry and mineralogy of dust is investigated. Snow-dust interactions based on a field campaign are also explained.

1.3 Outline of the dissertation

This doctoral study is based on the results presented in eight publications in peer-reviewed international journals, three of which are under review. In addition to this introductory section, the synthesis of the publications is presented in four chapters, followed by a section with conclusions and suggestions for future research. Two of the chapters also include results presented in abstracts from the European Geoscience Union (EGU) conference (Appendix A1, A2).

1.3.1 Scientific papers

Paper I. Dagsson-Waldhauserova, P., Arnalds, O., Olafsson, H., 2014a. Long-term variability of dust events in Iceland. *Atmospheric Chemistry and Physics Discussions* 14, 17331-17358. doi:10.5194/acpd-14-17331-2014.

Paper II. Dagsson-Waldhauserova, P., Arnalds, O., Olafsson, H., Skrabalova, L., Sigurdardottir, G. M., Branis, M., Hladil, J., Skala, R., Navratil, T., Chadimova, L., von Lowis of Menar, S., Thorsteinsson, T., Carlsen, H.K., Jonsdottir, I., 2014b. Physical properties of suspended dust during moist and low-wind conditions in Iceland. *Icelandic Agricultural Sciences* 27, 25-39.

Paper III. Meinander, O., Kontu, A., Virkkula, A., Arola, A., Backman, L., Dagsson-Waldhauserová, P., Järvinen, O., Manninen, T., Svensson, J., de Leeuw, G., and Leppäranta, M., 2014. Brief Communication: Light-absorbing impurities can reduce the density of melting snow. *The Cryosphere* 8, 991-995.

Paper IV. Arnalds, O., Olafsson, H., Dagsson-Waldhauserova, P., 2014. Quantification of iron rich volcanogenic dust emissions and ocean deposition from Icelandic dust sources. *Biogeosciences Discussions* 11, 5941-5967.

Paper V. Dagsson-Waldhauserova, P., Arnalds, O., Olafsson, H., 2013a. Long-term frequency and characteristics of dust storm events in Northeast Iceland (1949-2011). *Atmospheric Environment* 77, 117-127.

Paper VI. Dagsson-Waldhauserova, P., Arnalds, O., Olafsson, H., 2013b. Long-term dust aerosol production from natural sources in Iceland. *Journal of the Air & Waste Management Association*, DOI:10.1080/10962247.2013.805703

Paper VII. Arnalds, O., Thorarinsdottir, E.F., Thorsson, J., Dagsson-Waldhauserova, P., and Agustsdottir, A.M., 2013. An extreme wind erosion event of the fresh Eyjafjallajökull 2010 volcanic ash. *Nature Scientific Reports* 3, 1257.

Paper VIII. Dagsson-Waldhauserova, P., Arnalds, O., Olafsson, H., Hladil, J., Skala, R., Navratil, T., Chadimova, L., Meinander, O., 2014. Snow-dust storm: A case study from Iceland, March 7th 2013. *Aeolian Research*. submitted.

1.3.2 Organisation of the thesis

The introductory section is followed by the Chapter 2 – “Variability and trends of dust events”. This chapter assesses the frequency and severity of dust events based on meteorological dust observations in Iceland since 1949, seasonal trends in dust production and geographical differences of dust climatology in Iceland. The section is based on the Papers I, V and VI.

Chapter 3, “Physical and chemical properties of dust”, describes ground and air-borne measurements of aerosols conducted in Iceland. In addition, laboratory chemical and mineralogical analyses of dust aerosol and sediments are introduced. Papers I, II, VIII and Abstract EGU I (Appendix A1) are contributing to this section.

“Recent volcanic inputs to dust variability” is the title of Chapter 4, which explains the role of freshly deposited volcanic ash on the dust frequency in Iceland. The Eyjafjallajökull eruption in 2010 is used as a case study here. Chapter 4 is mainly based on the Paper VII and Papers I and V. Chapter 5 is titled “Dust deposition and climate aspects” and focuses on Icelandic dust deposition on a larger scale (ocean deposition, long range transport of dust particles). Here our research team quantifies the amounts of dust deposited over the oceans and glaciers. These estimates are supplemented with the dust-on-snow study revealing the albedo and snow melting changes as well as dust-on-snow deposition processes. A snow dust field campaign allows us to introduce first results on the optical properties of Icelandic dust which are compared to other absorbing aerosols. Chapter 5 summarizes the results from the Papers III, IV, VIII and Abstract EGU II (Appendix A2). Table 1.1 depicts the contribution of the papers to the chapters.

Table 1.1 Contribution of the papers to the dissertation chapters.

| Paper | Chapter 2 | Chapter 3 | Chapter 4 | Chapter 5 |
|--------------|------------------|------------------|------------------|------------------|
| I | X | X | X | |
| II | | X | | |
| III | | | | X |
| IV | | | | X |
| V | X | | X | |
| VI | X | | | |
| VII | | | X | |
| VIII | | X | | X |

The papers III, IV and VII are included in this dissertation despite being co-authored by the author of this thesis. Paper III is based on the field campaign on soot deposition on snow in Finland in 2013. I personally participated in the campaign and proposed the idea of expanding the experiment of volcanic dust. Paper IV originates partially from the calculations in the papers I and V, comparing the first estimations of the dust production in Iceland. Paper VII is a case study where the calculations and field work were done by the author of this thesis.

2 Variability and trends of dust events

2.1 Introduction

Frequency of dust episodes is monitored in vicinity of many major desert areas of the world. The long-term dust variability studies are based on meteorological observations and present up to 90 years records from North America, Africa, Asia and Australia (N'TchayiMbourou et al., 1997; Qian et al., 2002; Natsagdorj et al., 2003; Ekström et al., 2004; Jamalizadeh et al., 2008; Steenburgh et al., 2012). The World Health Organization considers that annual $\text{PM}_{2.5}$ concentration of $10 \mu\text{g m}^{-3}$ and estimated visibility 67 km indicates health risk, or daily standard of $35 \mu\text{g m}^{-3}$ and visibility range 31 km (WHO, 2005). In comparison, visual range can be over 300 km in dry climates and 100 km in humid climates on clear days (Hyslop, 2009). Observations of visibility during dust events are a key indicator of the severity of dust events where no aerosol measurements are conducted. Wang et al. (2008) found a good correlation between PM_{10} concentrations and visibility during dust observations.

The Arctic regions can also produce high amounts of dust with the highest dust emissions associated with summer and early autumn (Nickling, 1978; Bullard, 2013). Dust concentrations in sub-Arctic regions peak in spring (April-June, Prospero et al., 2012). Contrarily, Ganopolski et al. (2009) calculated higher glaciogenic dust deposition in cold and winter periods than warm periods. Iceland is located at the border between the Arctic and sub-Arctic regions. The long-term frequency, the climatology and character of dust production have not been investigated in Iceland. Moreover, the relationship between dust observations, available dust concentrations and visibility during dust observation is not known for Iceland.

2.2 Methods

A network of 30 weather stations (15 in S Iceland, 8 in NE Iceland, and 7 in NW Iceland) operated by the Icelandic Meteorological Office (IMO) is used in the study. The majority of stations have been in operation at least since 1949. The data consist of conventional meteorological parameters such as wind velocity, wind direction, temperature and visibility, accompanied by synoptic codes of present weather. Present weather refers to atmospheric phenomena occurring at the time of observation, or which has occurred preceding the time of observation (IMO, 1981). The synoptic codes (ww) for present weather which refer to dust observation are 7-9, and 30-35. In addition, codes 4-6 are considered, but only if the codes for primary or secondary past weather (ww1, ww2) are 3 for blowing soil, dust, sand and dust storm (IMO, 1981). Weather observations are made 3-8 times a day.

Meteorological observations (synoptic codes for dust including dust codes 04-06 and visibility) were evaluated with available PM mass concentrations from the Environmental Agency of Iceland (EAI) and daily dust concentrations from Storhofdi, Vestmannaeyjar (Westmann Islands, Prospero et al., 2012). Most of the conventional dust studies abroad do not include synoptic codes 04-06 for “Visibility reduced by volcanic ashes”, “Dust haze” and “Widespread dust in suspension in the air” into the criteria for dust observation, hence the unique volcanic conditions of Icelandic dust areas. Comparing these codes with dust concentrations shows that PM is increased in about 80% of the 04-06 code cases. These codes are, however, not included in this long-term study.

The initial dataset is built from the occurrence of “dust observation” made at one or more weather stations. Long-term dust activity is expressed in dust days. A “dust day” is defined as a day when at least one station records at least one dust observation. There are no *in situ* dust measurements conducted in NE Iceland and only few stations in the southern part of Iceland provide PM concentrations for a short-time period. Therefore, we applied empirical relationships to estimate dust concentration from the visibility (D’Almeida, 1986; Wang et al., 2008; Leys et al., 2011). Dust events are also classified from visibility ranges based on criteria provided by Leys et al. (2011) and Wang et al. (2008). For the PM stations, we apply a power regression to determine the relationship between dust concentrations and visibility during dust codes, including 04-06 (methods detailed in Wang et al., 2008).

2.3 Results and discussion

A mean of 34.4 dust days per year was observed in Iceland during the period 1949-2011. Such frequency is comparable to dust frequencies in the active parts of China (Qian et al., 2002), Mongolia (Natsagdorj et al., 2003), and Iran (Jamalizadeh et al., 2008). However, synoptic codes 04-06 (not included in dust studies abroad) showed a good agreement with increased PM₁₀ concentrations in Iceland. Including these codes into the criteria for dust observation, the annual mean dust-day frequency was 135 dust days (101 dust days in S Iceland, 34 dust days in NE Iceland). Such frequency can be found in parts of Australia and Africa (Ekström et al., 2004; N’TchayiMbourou et al., 1997). Trends in global dust emissions show high dust frequency during the 1950-1960s and low frequency during 1980s in the USA, Australia and China as well as in Iceland (Qian et al., 2002; Ekström et al., 2004; Steenburgh et al., 2012). The 2000s were reported as the most active decade in NE Iceland and Iran (Jamalizadeh et al., 2008).

The atmospheric surface pressure field determines whether dust plumes travel in a north-easterly or southerly direction. An annual mean of 16.4 dust days (total of 1033 days) was recorded in NE Iceland and about 17.9 dust days (total of 1153 days) occurred annually in S Iceland. The Arctic dust events (NE Iceland) were typically warm and during summer/autumn (May-September) while the sub-Arctic dust events (S Iceland) were mainly cold and during winter/spring (March-May). About half of the dust events in S Iceland occurred in winter or at sub-zero temperatures. The Arctic dusts correspond to the seasonality found in Alaska (Nickling, 1978; Bullard, 2013), while the cold winter/spring dusts can be found in Mongolia (Natsagdorj et al., 2003). Dust events (DE) in NE Iceland (Arctic DE) were warmer ($T_{\text{mean}}=10.5^{\circ}\text{C}$) and with lower wind velocities ($WS_{\text{mean}}=10.3 \text{ ms}^{-1}$) than sub-Arctic DE in S Iceland ($T_{\text{mean}}=3^{\circ}\text{C}$, $WS_{\text{mean}}=13.6 \text{ ms}^{-1}$). The most common DE

wind directions in NE Iceland were SW-S and SSE-SE, while the N-NE winds were predominant for S Iceland.

The Grimsstadir station (NE) is the dustiest weather observation location in Iceland with > 12 dust days annually. The stations, reporting high dust frequency, are also Hofn (S), Vatnsskardsholar (S), Egilsstadir (NE), and Hella (S). The stations in S Iceland reported higher frequency and severity of DE (low visibility and high wind speeds) than stations in the NE. This is likely due to the close proximity of the S stations to the dust sources as well as more stations are located in the S. The local dust sources in S Iceland are affected by milder oceanic climate during the winter while the NE highland dust sources are covered with snow for much of the winter. There were 32 “Severe Dust Storms” (visibility < 500 m) observed in Iceland (14 in NE mostly in the 1950s, 18 in S mostly in the 2000s). A good correlation was found between PM₁₀ concentrations and visibility during dust observations at the stations Vik and Storhofdi.

This study confirms that polar deserts in the Arctic and sub-Arctic region are important contributors to the global dust cycle as suggested in Bulard (2013). Dust in Iceland is emitted the year-round and often in winter or at subzero temperatures. It can be concluded that Iceland is among the dustiest areas of the world and likely the largest and the most active dust source in the Arctic/sub-Arctic region.

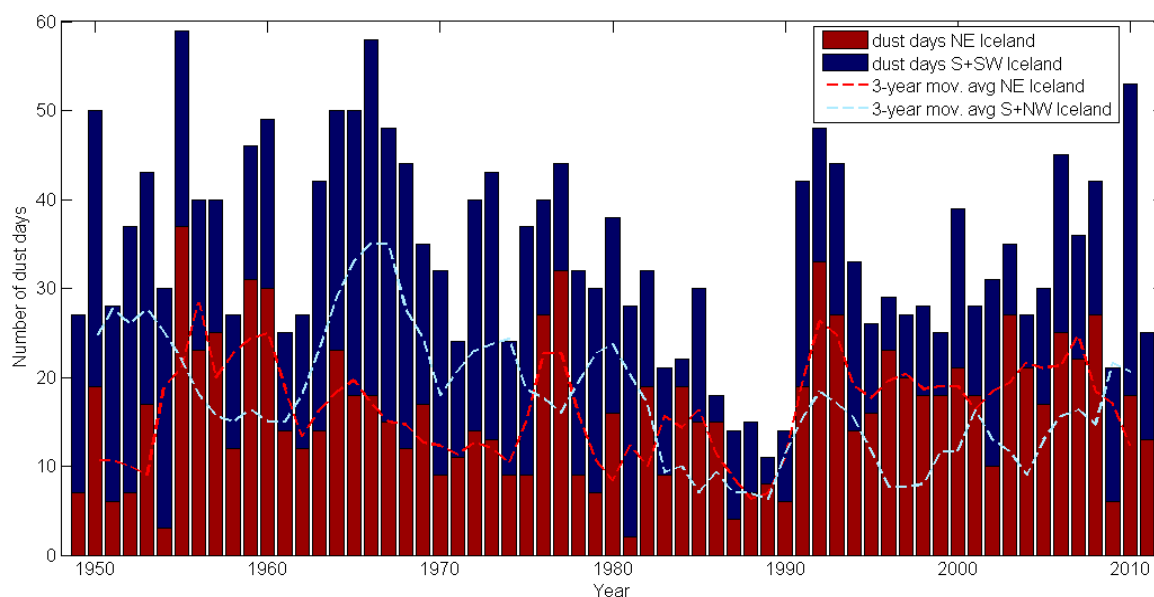


Figure 2.1 Number of dust days (blue bars for southern and north-western part of Iceland, brown bars for Northeast Iceland) and 3-year moving averages of dust day frequency (red for NE, light blue for S Iceland).

3 Physical and chemical properties of dust

3.1 Introduction

Dust activity in the major desert areas is monitored. The data on dust concentrations are provided in real time and dust weather forecasts are generated by using several models (NAMEE, 2014). Dust measurements within cold and high-latitude regions are mainly seasonal and employ measurement techniques focused mostly on the coarser silt-sized or sand-sized particles (Nickling, 1978; Arnalds et al., 2001; Bullard, 2013). Air particle monitoring using a range of automatic instruments is conducted far from the major dust sources in the Arctic (NILU, 2013). Dust forecasting and models require relevant information on physical characteristics of aeolian materials, such as chemistry, mineralogy, PM mass and number concentrations. Icelandic dust is of volcanic origin and is of different nature compared to crustal dust. Therefore the knowledge of the physical properties of volcanic dust is needed for dust parameterizations of the dust models.

Measurements of particle mass concentrations in Iceland have primarily been made in relation to the Eyjafjallajökull eruption in 2010 (Leadbetter et al., 2012) or areas distal from the dust sources (Thorsteinsson et al., 2011; Blechschmidt et al., 2012). No direct measurements of dust concentrations have been made within the major dust sources in Iceland. Size segregated particle mass concentrations and number-size distributions (number of particles in defined particle size ranges) of dust aerosol can provide a better understanding of physical properties, such as textural, morphological and shape characteristics, and the possible health impacts of dust events (Harrison and Yin, 2000; Morman and Plumlee, 2013). Studies on number concentrations which include particles $<10\text{ }\mu\text{m}$ have not been published in the literature to date for Icelandic conditions.

The main aim of this research is to provide an overview of physical properties of Icelandic dust, as exemplified by dust from Maelifellssandur and Skeidararsandur, which are two of Iceland's main dust sources. Results of the first synchronized measurements of particle number and mass concentrations during a dust event directly within a dust sources in Iceland are reported. This information is combined with mineralogical and geochemical analyses of the source material. Cases of vertical stratification and aerosol composition of the sub-Arctic atmosphere up to 16 km, measured after a season of dust episodes are also studied in detail.

3.2 Methods

3.2.1 Atmospheric dust measurements

Investigations of aerosol physical properties in Iceland were conducted both on ground and via air borne measurements. Atmospheric Dust Measurements in Iceland (ADMI 2013) was a pioneering project to investigate the physical characteristics of dust aerosol *in situ* at the dust source. The ADMI 2013 took place in southern and south-western Iceland on 8-18 August 2013, a period of high precipitation and low winds. We employed two TSI 8520 DustTrak Aerosol Monitors (DustTrak) and one TSI Optical Particle Sizer 3330 (OPS) placed on the glacial floodplain Maelifellssandur. The DustTrak is a light-scattering laser photometer measuring aerosol mass concentrations for particles ranging in size from 0.1 to 10 μm . The OPS provides particle concentration and particle size distribution measurements using single particle counting technology, for particles with optical diameters from 0.3 to 10 μm . It employs optical scattering from single particles where particle pulses are sized and binned in up to 16 different channels. In addition to these measurements, mineralogical and chemical analyses were made for the sample taken from the active surface layer during the dust event (see section 3.2.2).

Three air-borne campaigns of the Light Optical Aerosols Counter (LOAC) were conducted with meteorological balloons in Iceland. LOAC uses a new optical design retrieving the size number concentrations in 19 size classes between 0.2 and 100 micrometers. It also provides an estimate of the main nature of aerosols (dust, Black Carbon, sulphuric acid, water droplets and ice). These campaigns were focused on aerosol distribution in vertical profile of atmosphere.

3.2.2 Laboratory measurements

The compositions of two dust samples from two dust events (tephra glass and mineral grains) were studied using backscattered electrons and quantitative x-ray analysis (EDX SEM) on samples fixed in resin and polished to planar cross-sectional surfaces. Major mineral compositions were also checked by x-ray powder diffraction (XRD).

3.3 Results and discussion

3.3.1 Atmospheric dust measurements

The ADMI resulted in one dust event at the dust source, which was measured under unique conditions, during low wind/windless conditions when the surface was still moist after previous precipitation. The main driver of dust suspension was direct solar radiation and consequent surface heating. Despite being a rather small dust event, high amounts of dust were passing the instruments and the particle number concentrations were extremely high. Maximum particle number concentration (PM_{~0.3-10} μm) reached 149,954 particles cm^{-3} per minute while mass concentration (PM_{<10} μm) was 1757 $\mu\text{g m}^{-3}$. Maximum concentrations of the PM_{2.5} fraction were measured as 85,528 particles cm^{-3} in number concentration with mass reaching 116 $\mu\text{g m}^{-3}$. Suspended glaciogenic dust was very fine

with the highest number of particles in the size range 0.3-0.337 μm , followed by particles 1.5-5 μm in diameter. Such high number concentrations of particles 0.3-10 μm have only been reported during a volcanic eruption (Vogel et al., 2012). Close-to-ultrafine particle size distributions showed a significant increase in number with the severity of the measured dust event (during dust peaks). Number concentrations were well correlated with mass concentrations. This is unusual because mass concentration increases with larger particles suspended, while the number of particles decreases. Such strong relationships as found here resulted likely from the moist soil conditions and high humidity. The glacial floodplain was still moist and therefore only fine, dried particles were uplifted. Dust suspension of fine moist particles during low wind/windless conditions was initiated in < 4 hours. The highest number concentrations for submicron particles are generally attributed to wind speeds < 2 ms^{-1} (Weber et al., 2006).

Icelandic dust events can be much more severe than the event reported above. There are > 6 dust events with visibility < 5 km (measured at some distance from the source) on average in Iceland each year. For example, PM_{10} mass concentrations during dust storm in March 2013 were measured as > 6500 $\mu\text{g m}^{-3} \text{ min}^{-1}$ while the mean (median) PM_{10} concentration during 24-hour storm was 1,281 (1,170) $\mu\text{g m}^{-3}$. Generally, severe dust storms in Iceland result in PM_{10} concentrations exceeding 7000 $\mu\text{g m}^{-3}$.

The extension of aerosol concentration measurements from the ground to stratosphere gives information on vertical stratification and aerosol composition of the sub-Arctic atmosphere. It was found that dust can stay in the atmosphere several days after the dust event, despite rainy and snowy conditions. The LOAC identified a thin layer of volcanic dust at an altitude of one km one day after the event. The particles were > 1 μm in this layer, suggesting that they were mixed inside a haze or small cloud layer.

Figure 3.1 shows that the nature of the aerosols changed with altitude during the flight conducted in Reykjavik in November 2013. Small liquid and sea salt particles were detected close to the surface while dust particles were in height about 1 km. The instrument passed cloud (probably mixture of liquid and ice particles at the height of about 4 km, and cirrus cloud at the height 6-8 km where icy particles were found. Background carbon particles were present at around 5 km altitude between the cloud layers. In the stratosphere above 12 km, reasonably high number concentrations of particles > 1 μm (even tens of micrometers) occurred showing unusual conditions. It might have been an extraterrestrial dust, as previously observed sometimes by *in situ* counting (Renard et al., 2005). For the smaller size, LOAC indicate mainly carbon particles, as previously observed in the stratosphere (Renard et al., 2008). This launch showed that aerosols such as Black Carbon can be found in the sub-Arctic atmosphere, far from their sources.

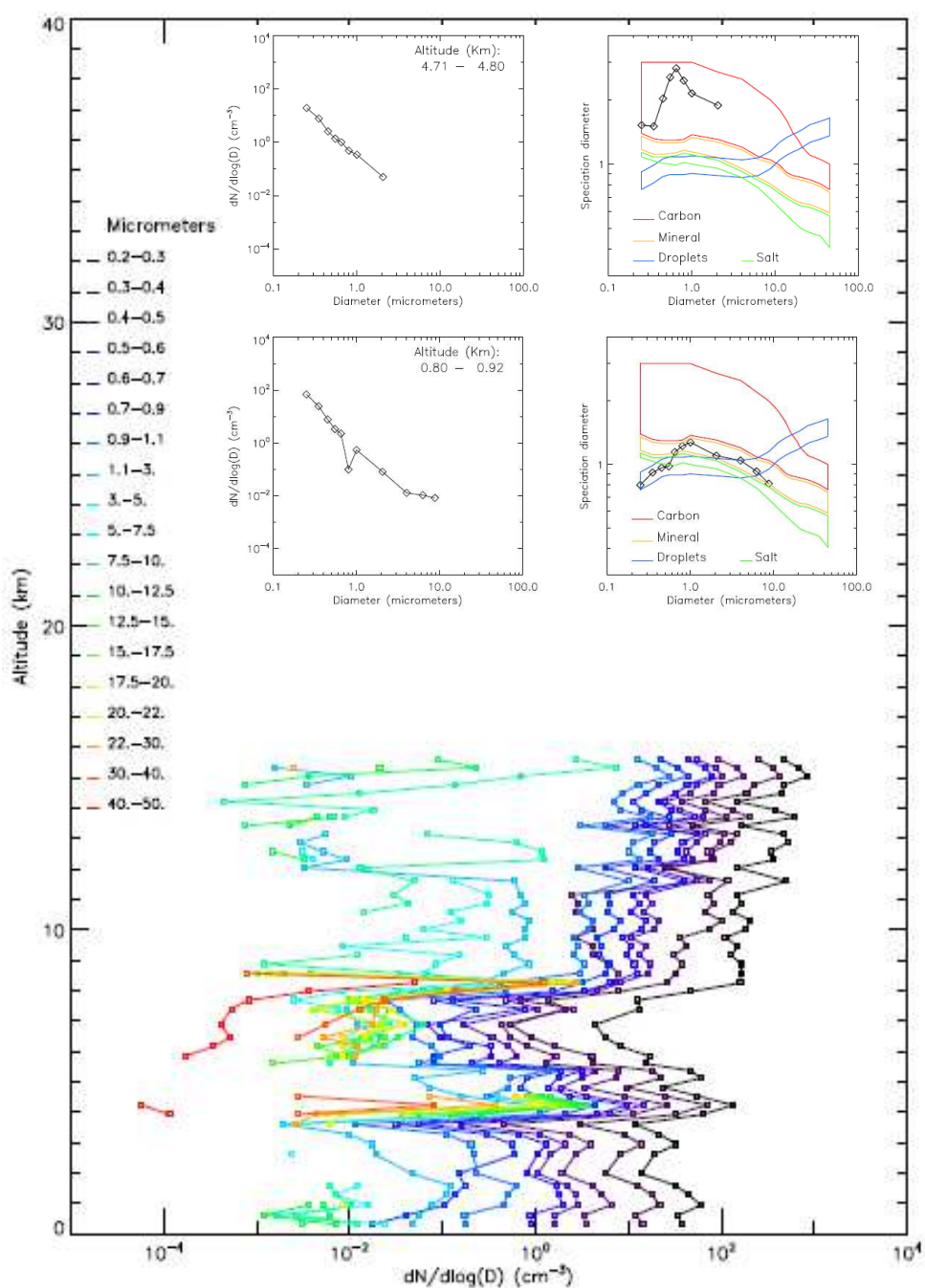


Figure 3.1 Vertical profile of particle number concentrations during the LOAC launch on November 7th, 2013 in Reykjavik, Iceland. Aerosol composition at two altitudes (about 1 km and 5 km) is to the right on top.

3.3.2 Mineralogical and geochemical analyses

Mineral grains and geochemical composition of Icelandic dust differs between the major dust sources (Baratoux et al., 2011; Oladottir et al., 2011). A sample from the active

surface layer during the dust event at one of the major dust sources, Maelifellsandur, was analyzed. Second dust sample was obtained from the dust layer deposited on snow during the severe dust storm in Reykjavik in March 2013. The origin was likely from the second major dust source, Skeidararsandur. Both samples included also traces of fresh volcanic material from the Eyjafjallajökull 2010 eruption and Grimsvotn 2011 eruption.

The mineralogy and geochemical composition showed that the glaciogenic dust contains sharp-tipped shards with bubbles and 75-80% of the particulate matter is volcanic glass rich in heavy metals. Major element composition shows significant depletion of SiO₂ in Icelandic dust compared to average crustal dust with about 58% SiO₂ reported by Weast et al. (1966). Table 3.1 summarizes the average element composition of two dust samples with unusually high amounts of FeO and TiO₂ contents. High heavy metal contents of these two examples are in contrast to composition of tephra glasses from recent eruptions and other sources in Iceland (Sigmarsson et al. 2011, Navratil et al., 2013). However, the depletion in silica is indicative for all Icelandic materials.

Table 3.1. The chemical composition of the Maelifellssandur and Skeidararsandur dust event samples based on EDX analyses of polished surfaces of individual glass particles.

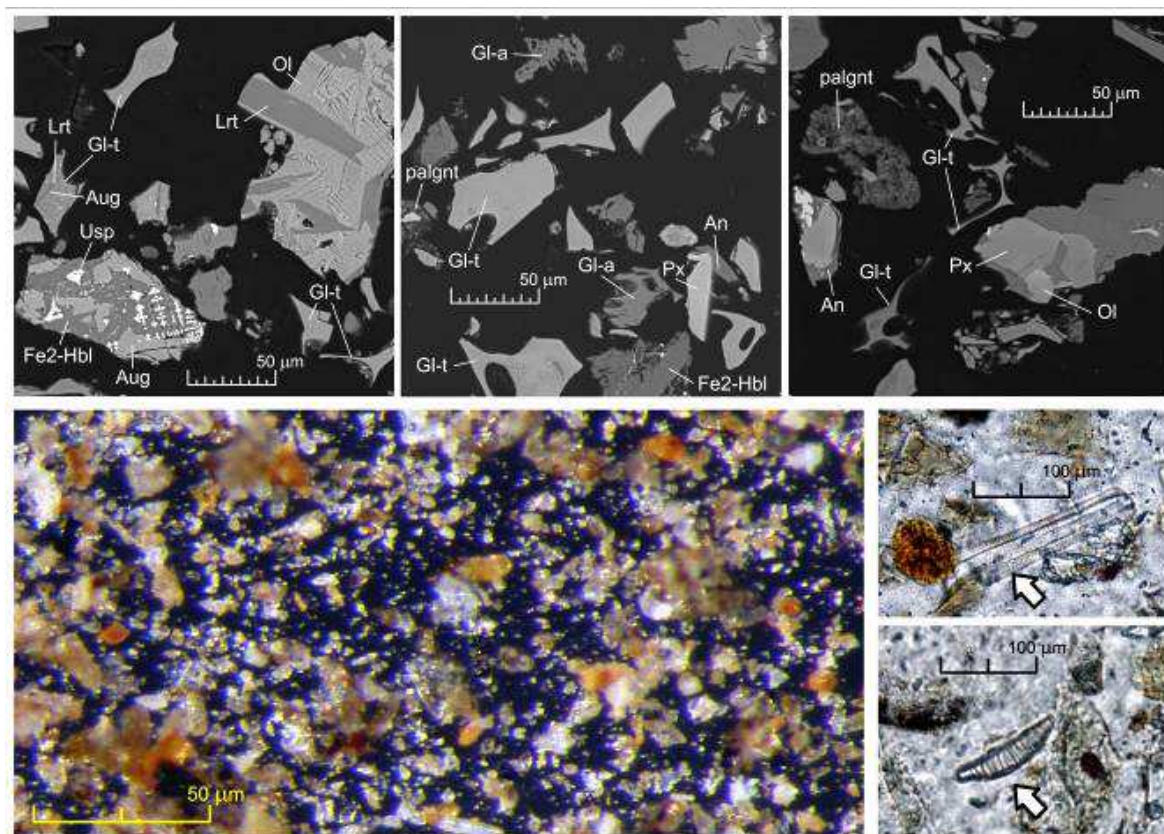
| Average values | SiO₂ | FeO | Al₂O₃ | CaO | TiO₂ | MgO | K₂O +Na₂O |
|-----------------------------|------------------------|------------|------------------------------------|------------|------------------------|------------|--|
| Maelifellssandur dust event | 42.57 | 17.00 | 14.20 | 11.55 | 5.61 | 4.94 | 4.14 |
| Skeidararsandur dust event | 45.00 | 14.50 | 14.50 | 12.00 | 3.50 | 6.25 | 4.00 |

Mineralogical analysis of the dust deposited on snow (second sample) revealed quite variable size range for the grains of 1–250 µm in diameter (average 17 µm). About 20% of particles were in range 10-50 µm and 10% were > 50 µm. A severe dust storm measured in 2010 (Chapter 4) transported mostly particles > 125 µm, with grains up to 8 mm. This shows that the PM₁₀ concentration measurements overlook a significant part of the suspended dust mass during dust storm events.

Glass morphology was characterized by numerous ~10–20 µm gas bubbles. Low frequency of bubbles corresponded to massive shards, but high frequency occurs in bubble-wall shards. The glass is extremely angular and of sharp-tipped shards (Figure 3.2). The shard-faces are often curved and concave. Rare alkali- and silica-rich glasses showed different, very fine pipe-vesicular structures (Figure 3.2, top-right). Such elongated shapes are more similar to asbestos particles or Black Carbon than mineral dust, and may pose health risks. Donaldson et al. (2006) found that particle length is a predictor for pathogenesis in fibrous particles, as known from asbestos. Moreover, high amount of bioavailable metals in the dust increases the inflammatory capacity of the PM, which may lead to negative health effects (Morman and Plumlee, 2013).

Diatoms and organic matter is also transported during the dust events in Iceland. We identified diatoms during the March 2013 event. *Rhopalodia* and *Epithemia* diatom species (likely epiphytic) are benthic and may be present in shallow pools or waters around the

edges of lakes and rivers (Figure 3.2). However, the identification of exact location from the 49 examined lakes and 139 diatom taxa found in Iceland is complicated (Karst-Riddoch et al., 2009).



*Figure 3.2 Microscopic images of the dust material. Upper row: Backscattered electron images of planar-polished sections of the dust particles; the analyzed and interpreted minerals and glasses are marked by symbols: An – andesine, Aug – augite, Fe2-Hbl – ferrohornblende, Lrt – labradorite, Ol – olivine, Px – pyroxene, Usp – ulvospinel; Gl-t – volcanic glass of tholeiitic series, Gl-a – volcanic glass of alkalic series; palgnt – palagonitic material. Lower row: The fine particle distributions are illustrated in the figure on the left (optical microscope, dark field). Examples of diatoms *Rhopalodia* sp., poss. *R. gibba* (upper) and *Epithemia* sp., poss. *E. adnata* (lower).*

4 Recent volcanic inputs to dust variability

4.1 Introduction

Volcanoes are one of Earth's most dramatic and violent agents of change. Volcanic eruptions can bury landscapes with tephra and create extensive areas with unstable surfaces (del Moral and Grishin, 1999). Wind erosion of unstable newly deposited volcanic ash has been reported to cause a range of problems such as burial of vegetation and agricultural land, and impact on livestock and humans (Wilson et al., 2011; Arnalds, 2013). Wind erosion of fresh volcanic deposits are believed to have caused rapid and large scale ecosystem destruction during historic times in Iceland, with volcanic sand materials encroaching on fully vegetated agricultural areas, leaving sandy deserts behind (Arnalds et al., 2001). Yet surface conditions during erosion events following volcanic eruptions have not been investigated to date.

Iceland is a volcanic island with over 30 active volcanic systems resulting in eruptions every 3-4 years on average (Gudmundsson et al., 2008). Recently, two volcanic eruptions occurred in Iceland – the Eyjafjallajökull eruption in 2010 and the Grimsvotn eruption in 2011. We monitored volcanic ash resuspension in vicinity of the Eyjafjallajökull volcano the first year following the eruption. The eruption started in spring of 2010 with a strong volcanic explosion when the major ash plume reached over 9 km height and about 155 km in distance from the volcano (Arason et al., 2011). Fine-grained volcanic ash particles were released into the atmosphere and dispersed globally (Stohl et al., 2011). Intense tephra discharge continued for several days and the Nordic Volcanological Institute reports about 100 million m³ of tephra ejected (Arason et al., 2011). The summer of 2010 was dry with frequent occurrence of high winds, resulting in numerous wind erosion events.

This chapter presents results from the *in situ* measurements of fresh volcanic ash events and resuspension following the eruption in 2010. In addition, we discuss the frequency of long-term meteorological dust observations during the “eruption” and “non-eruption” years.

4.2 Methods

Wind erosion was monitored on a 1200 m long transect across the main ash deposition lobe about 10 km from the crater of Eyjafjallajökull. The area received 2-5 cm of volcanic ash during the eruption (Figure 4.1). The amount of windblown material was estimated at five locations on the transect. We employed two methods to determine aeolian sediment transport rates: sediment accumulation in dust traps and an electronic saltation sensor. The BSNE wind erosion samplers developed by Fryrear (1986) were placed at several locations,

30 cm and 60 cm above the surface. In addition, there was one set of five samplers placed at 10, 30, 60, 90, and 120 cm height at the same location.

The automated wind erosion counter (SENSIT) is an instrument programmed to give a pulse upon an impact from moving grains using piezo-electric material. These pulses are magnified and counted. An automatic weather station measured simultaneously wind velocity and other parameters with impact counts, allowing for relating wind speeds and wind erosion intensity. The samplers give a better overview of total particle movement (erosion), while the data generated by the automatic sensor are ideal to study the characteristics of each storm. The dust traps were emptied after each storm period. A characteristic height distribution curve for each location obtained by the set of five samplers placed at different heights was used to calculate transport for each period where only one sampler was in place (Arnalds et al., 2012). The unit of measurement is amount of material transported over 1 m wide transect (kg/m) in total or per unit time (kg/m/hr; kg/m/minute).

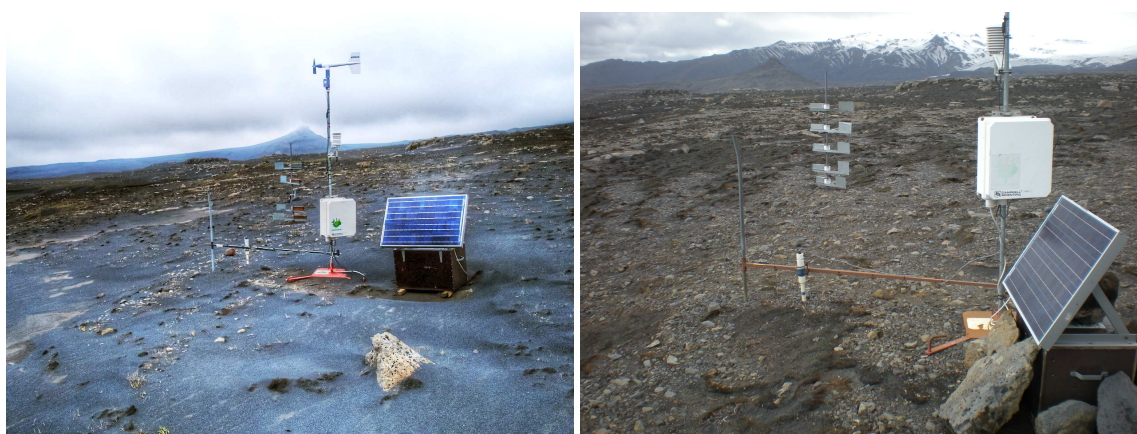


Figure 4.1 Research site at Eyjafjallajökull during the year of the eruption in 2010 (left, ©Johann Thorsson) and one year after in 2011 (right, ©Pavla Dagsson-Waldhauserová). Majority of the fresh volcanic material was removed during extreme wind erosion events in 2010.

An annual frequency of the meteorological dust observations (see Chapter 2) during the years following the eruption was compared to the years when no eruption occurred. This correlation is to evaluate the impact of volcanic eruption on the long-term dust event frequency.

4.3 Results and discussion

The Eyjafjallajökull eruption was declared finished in June 2010. We measured > 30 wind erosion events at Eyjafjallajökull research site during the period June – October 2010. This

result for a period of five months at one location is higher than the long-term annual average of 34 dust days for the whole Iceland. However, conventional meteorological dust observations from the surrounding weather stations (Vatnsskardsholar, Vik, Kirkjubaejarklaustur, Storchofdi) did not report > 8 dust days in 2010. Adding codes 04-06 among the conventional dust codes (see Chapter 2), the frequency of dust days (DD) resulted in 27 DD in Kirkjubaejarklaustur, 42 DD in Vatnsskardsholar, and almost 60 DD in Storchofdi in 2010. This strongly indicates that also 04-06 codes should be included in the long-term dust studies in Iceland. Generally, the volcanic ash deposited during the 2010 Eyjafjallajokull caused serious dust storms in the southern part of Iceland (Schumann et al., 2011; Leadbetter et al., 2012; Petersen et al., 2012) as confirmed by the meteorological dust observations (Figure 2.1), but no increase was recorded in dust activity in NE Iceland. Contrarily, the eruption of Grimsvotn in 2011 had almost no influence on dust day frequency in Iceland. Similarly, no relationship has been found between dust day frequency and eruption inputs in the recent past (1949-2001). This suggests that the meteorological conditions (mainly winds) are ruling the dust occurrence in Iceland, but some eruption inputs are also important, generating temporary spikes in dust activity. The tephra produced from the individual volcanic systems in Iceland differs in composition and physical properties. Deposited tephra after the Hekla eruptions is coarser and does not cause so many volcanic ash resuspension events like the fine tephra from the Eyjafjallajokull.

The Eyjafjallajokull eruption in Iceland in 2010 provided a unique opportunity to study volcanic ash transport in natural environment. Thirty captured volcanic ash/dust events were of different severity, meteorological conditions and particle composition. In September 2010, an extreme storm was recorded with the maximum wind speed of 38.7 ms⁻¹. The maximum saltation was 6825 pulses per minute, which is the highest we have measured during the 16 years of operation of the instrumentation at various locations. Surface aeolian transport over one m wide transect and up to 150 cm height reached 11,800 kg. The largest previously measured amount in Iceland in one storm was about 4,200 kg m⁻¹ at Landeyjasandur, South Iceland, in 2004 (Agustsdottir, 2007). Griffin et al. (2002) estimates that a single large dust storm can deliver more than 200 tons. Our results of 11 t per meter gives significantly higher number for total extent of the dust storm. We have not been able to find such high measured transportation rates in the published literature in other countries. This storm is therefore among the most extreme wind erosion events recorded on Earth.

The mean grain size of the suspended particles from all events ranged from 0.13 to 0.69 mm, and grains > 2 mm were moved during the most intense storms (Figure 4.2). This is coarser than reported for wind erosion within other active aeolian areas (Pye and Tsoar, 1990). The vertical particle size analysis showed that 90% of the material mass was in the saltation layer up to 45 cm above surface during the moderate events, while the saltation layer increased to 85 cm above surface during the severe storms. Such height of saltation layer is considerably higher than the 20–40 cm height reported elsewhere (van Donk and Skidmore, 2001; Zobeck, 2003). Moreover, smaller particles of < 0.125 mm were almost absent in samplers at all heights after severe storms. The sorting of surface materials likely takes part during the early storms with the fine particles being lost into the atmosphere while larger particles still remain at the sites. Mean wind velocity during observed dust events increased from about 10 to 18 ms⁻¹ from June to October. The saltation increased exponentially with average wind speed to the saturation point and then grew gradually. The

saltation saturation point in natural environment near Eyjafjallajokull is at about 24 ms^{-1} wind speed and 5 500 pulses per minute saltation.

The Eyjafjallajokull wind erosion storms caused dust emissions extending several hundred km from the volcano affecting both air quality (Thorsteinsson et al., 2012) and ecosystems, showing how wind erosion of freshly deposited ash prolongs impacts of volcanic eruptions. Fresh volcanic material is, however, relatively rapidly removed from the environment (Figure 4.1). The chemical and mineralogical analyses presented in Chapter 3 showed only a small portion of Eyjafjallajokull and Grimsvotn volcanic material that was found in the samples from the eruption areas about 2-3 years after the eruptions.

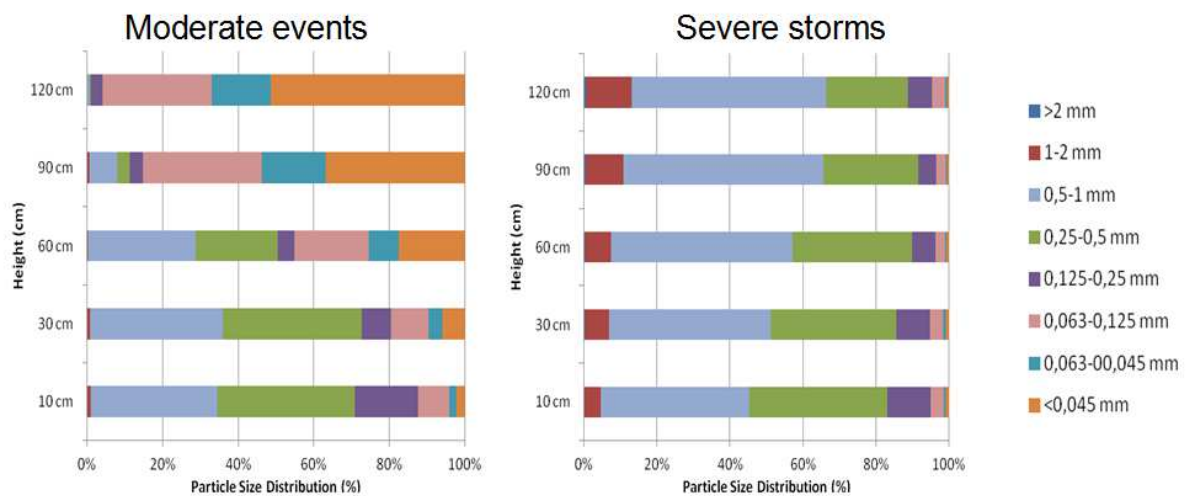


Figure 4.2 Vertical particle size distributions during the moderate and severe wind erosion events at Eyjafjallajokull. Particles $< 0.125 \text{ mm}$ were almost not found in samplers at all heights after severe storms, while the proportion of grains $> 1 \text{ mm}$ increased with the height.

5 Dust deposition and climate aspects

5.1 Introduction

Suspended dust aerosols play an important role in the atmosphere-ocean climate system. Global estimates of mean annual dust emissions range from 500 to 5000 million tons per year with the global oceans estimated to receive 300-500 million tons (Engelstaedter et al., 2006). Commonly referenced is that Sahara contributes about 200 million tons per year to the tropospheric aerosol budget (Morales, 1979). Dust in high amounts is transported long distances. About 13 million tons of African sediment travel thousands of kilometers to the South American continent every year (Griffin et al., 2002). African and Asian dust particles have also been identified in the Arctic glaciers (Drab et al., 2002).

Some Icelandic dust plumes detected on satellite images reach > 1000 km distance over the Atlantic Ocean and Arctic Ocean (Arnalds, 2010). Drab et al. (2002) found Icelandic particles in the ice-cores of Greenland's glacier. Volcanic ash particles after eruptions were detected thousands of kilometres from Iceland (Rose et al., 2006, Navratil et al., 2013). The majority of Icelandic dust is, however, deposited on land and the marine areas around Iceland. Prospero et al. (2012) suggested that Icelandic dust emissions could contribute a substantial amount of iron to the North Atlantic. Distribution of fresh volcanic material resulted in significantly elevated iron levels south of Iceland during the 2010 Eyjafjallajökull eruption (Achtenberg et al., 2013). Volcanic ash is commonly subjected to intense aeolian redistribution (see Chapter 4), while Icelandic glaciogenic dust sources are composed of iron rich volcanic deposits (see Chapter 3). In spite of first calculations of dust day frequency in Iceland (Chapter 2), little is known how much volcanic material is blown to the oceans around Iceland.

Large areas in Iceland and in proximity of Iceland are covered with glaciers, sea ice and snow. Dust events in southern part of Iceland occur often during winter or at sub-zero temperatures, resulting in snow mixed with dust or covered by a dust layer (Chapter 2, 3). Darker snow reduces snow albedo and accelerates snowmelt (Painter et al., 2012; Steenburgh et al., 2012). The effects of Icelandic dust deposited on snow are not known. Moreover, optical properties of Icelandic dust aerosol have not been investigated. Generally, direct radiative forcing of mineral dust is calculated as negative in the IPCC report (IPCC, 2013). In terms of climate forcing, Black Carbon has been found as the most powerful absorbing aerosol and the second most important human emission, only carbon dioxide is estimated to have a greater forcing (Bond et al. 2013). Icelandic volcanic dust is dark in colour and the optical properties might have more similarities with Black Carbon than with crustal dust.

5.2 Methods

5.2.1 Quantification of iron-rich dust deposition

We used two independent methods to quantify dust emissions from Iceland and dust deposition on sea. Firstly, aerial extent (map) of deposition on land was extended to ocean areas around Iceland. Secondly, number of dust events over the past decades (Chapter 2) and calculations of emissions and sea deposition for the dust storms were made.

A GIS-based dust deposition map for Iceland was presented by Arnalds in 2010. The map is based on soil metadata showing thicknesses between tephra layers (volcanic ash) of known age, main dry winds from each major dust source and landscape parameters downwind from the sources. We have extended the map to oceanic areas and included categories for very low deposition furthest away and extreme deposition closest to the aeolian sources. The aerial distribution of the deposition over sea is based on satellite images (MODIS; Aqua and Terra) taken over the past decade. Most of the dust is expected to settle relatively close to the source with a logarithmic drop in sedimentation with distance from the source (Figure 5.1).

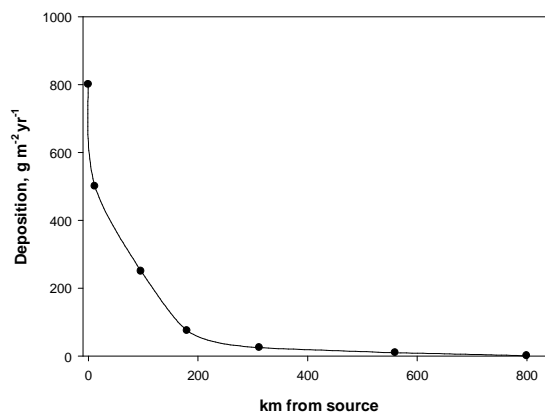


Figure 5.1 The logarithmic drop in sedimentation with distance from the sediment source; transect south from the dust sources at the southern tip of Iceland. The transect from Dyngjúsandur in Northeast Iceland has x-axis scale approximately divided by 2 (based on the area of the dust sources and dust frequency).

The second method is based on the meteorological dust observations, which are reported together with visibility (see Chapter 2). Visibility is an important indicator of dust event severity where dust concentration measurements are not available. The concentration of dust is based on: i) observed visibility at manned weather stations; ii) the horizontal extension of the dust plumes; and iii) the representativeness of the visibility observations are estimated from MODIS satellite images (example such as Figure 5.6). The winds in the atmospheric boundary-layer are estimated from ground-based and air-borne observations (upper air) as well as numerical simulation, and the thickness of the boundary-layer is estimated from the upper air observations and numerical simulations. The upper-air

observations are made at 00 and 12 UTC at Keflavik, SW Iceland and Egilsstaðir, NE Iceland. The numerical model, Harmonie (based on Arome, see Seity et al., 2011) was run with subgrid 1-D turbulence scheme based on Cuxart et al. (2000) with a horizontal resolution of 2.5 km. The simulations are based on initial and boundary-conditions from the operational suite of the ECMWF. Four dust-storms with different severity (minor, medium, major) were selected for estimation of the total transport of suspended dust. In all four storms, the visibility in the dust plume was recorded, and the vertical, horizontal and temporal extension of the plume was estimated from the available data. The annual mean number of dust events (minor, medium, major) for S and NE Iceland was then used for the quantification.

Finally, the iron content of the dust was selected as an average of 10% Fe of volcanic dust based on the common range for iron in Icelandic volcanic rocks reported from 6.5 to 12.5% (see also Chapter 3). It can be inferred that the total iron deposition to the oceans is about 1/10 of the weight of the dust load. An estimate of bioavailable Fe in the sea is based on evidence presented by Achterberg et al. (2013) after the 2010 Eyjafjallajökull eruption.

5.2.2 The Soot on Snow (SoS-2013) project

The Soot on the Snow (SoS-2013) experiment was carried out in Sodankylä (67°22'N, 26°39'E), Finland, to study the effects of deposition of Black Carbon (BC) and Icelandic dust on the surface albedo, snow properties and melt of the seasonal snow. Two dust samples were artificially applied on the snow surface to identify the effects of dust on snow pack - coarse dark volcanic sand and fine light-brown glaciogenic silt from the Myrdalssandur dust source. Other spots contained BC from wood burning chimneys and oil burners, while one spot was left as a reference clean-snow spot. The spectral reflectance of melting snow and dust in laboratory was measured using the Analytical Spectral Device (ASD) Spectrometer for 325-1075 nm. Moreover, laboratory and snow spectropolarimetric measurements were made using the Finnish Geodetic Institute Field Goniospectrometer FIGIFIGO (<http://www.polarisation.eu/index.php/list-of-instruments/view-submission/172>). This instrument uses multiangular reflectance to measure the albedo, polarization, and other snow properties. Spectral reflectance of snow was measured first days of the deposition (FIGIFIGO) and two weeks after the deposition (ASD).

5.3 Results and discussion

5.3.1 Iron-rich dust deposition from Icelandic dust sources

The total dust emissions range from 30.5 (dust event based calculation) to 40.1 million tons (GIS-map calculation), which places Iceland among the highly active dust sources on Earth. Two different values reflecting this range are obtained by independent methods, but are, however, in relatively good agreement. The dust emissions in Iceland presented here are of the order of 0.6-7.2% of the total estimate for global dust emissions of 500 to 5000 million tons given by Engelstaedter et al. (2006). They noted North Africa is by far the largest source of dust with 170–1600 million t, but our numbers are 1.9–21% of these

estimates of North African dust. Icelandic dust emissions are 15-20% of the Saharan estimate of 200 million t given in Morales (1979).

Ocean deposition of dust ranges between 5.5 (dust events calculations) and 13.8 million tons (GIS-map calculation). The dust deposition per unit area west of the Sahara is considered to be about $10 \text{ t km}^{-2} \text{ yr}^{-1}$ (Duce et al., 1991). The corresponding average number from the data presented here is $10.4\text{--}25.7 \text{ t km}^{-2} \text{ yr}^{-1}$ on average over $370,000 \text{ km}^2$ sea area, equal to or substantially greater than the rates reported by Duce et al. (1991). Calculated iron deposition from the Icelandic dust ranges between 0.56 and 1.4 million tons distributed over wide sea areas. Jickells et al. (2005) indicated that atmospheric sources of iron were of the order of 16 million tons Fe per year. The Icelandic iron deposition to oceans is a sizeable proportion of the estimate of the global total (3.5 – 8.7%). Only part of the iron in the dust/ash becomes bioavailable. A range of Fe

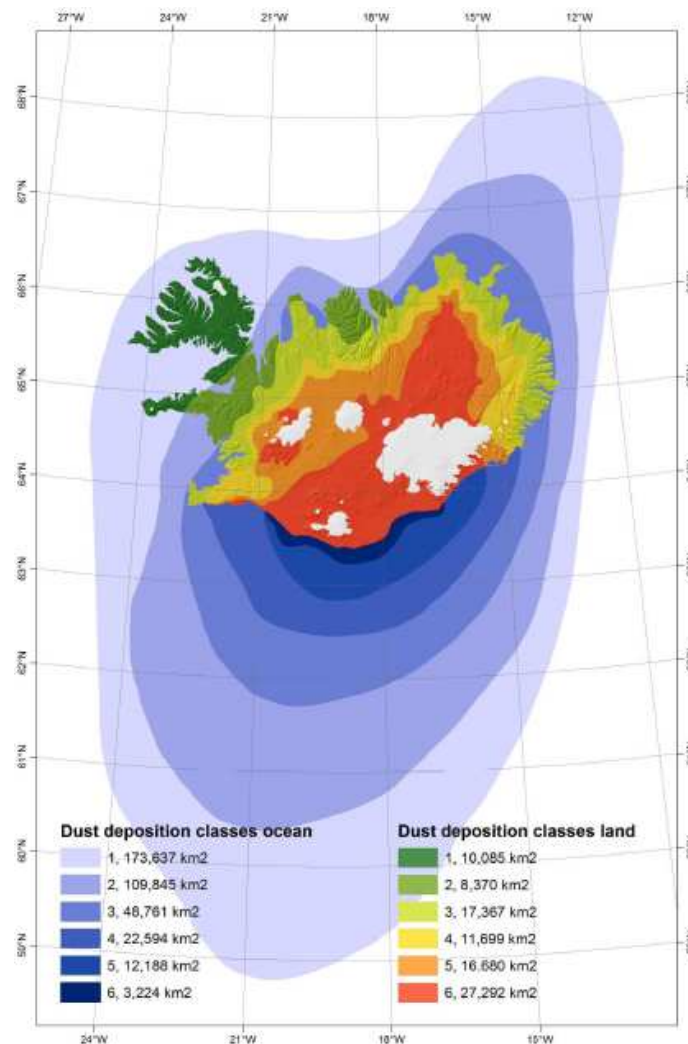


Figure 5.2 Average distributions of dust and volcanic ash around Iceland. Deposition is split in six dust deposition categories (category 1 - deposition 5 t km^{-2} , 2 - 20 t km^{-2} , 3 - 44 t km^{-2} , 4 - 119 t km^{-2} , 5 - 350 t km^{-2} , 6 - 500 t km^{-2}), and is an extension of previously published map for terrestrial Iceland (Arnalds, 2010).

bioavailability has been reported in the literature with 0.004 - 0.04 % (Olgun et al., 2011). We made an effort of quantifying bioavailable iron based on 0.02% bioavailability (Achterberg et al., 2013), which shows that bioavailable Fe from dust sources are of the order 0.04 to $> 10 \text{ mg m}^{-2} \text{ yr}^{-1}$. However, fresh volcanic ash releases more iron than several years old volcanic dust (Jones and Gislason, 2008). Volcanic ash from the Eyjafjallajökull eruption consists of less iron than about hundred years old tephra from the Katla eruption (see Chapter 3). These are, however, the first quantitative estimates of total dust emissions and oceanic deposition from Iceland. Iron is a limiting nutrient for primary production in the oceans around Iceland and the dust is likely to affect Fe levels in Icelandic ocean waters as suggested by Prospero et al (2012).

The dust deposition rates are depicted in Figure 5.2. The land areas including glaciers are receiving 25-26 million tons of dust per year based on both methods. Most of the Icelandic glaciers are located in proximity of the dust sources and the mean deposition on glaciers was estimated as $400 \text{ g m}^{-2} \text{ yr}^{-1}$ from the deposition map. It means that the deposition on the Icelandic glaciers is 4.5 million tons annually. The effects of dust deposition on snow albedo and melting rates are discussed in the following chapter (Chapter 5.3.2).

5.3.2 Impacts of volcanic dust deposition on snow

Two dust samples from Iceland were investigated on snow, volcanic sand and glaciogenic silt. The snow albedo during the time of the deposition was the highest for clean snow and slightly reduced for snow with volcanic sand. The soot reduced the snow reflectance significantly, but fine Icelandic silt reduced the snow reflectance the most, approximately

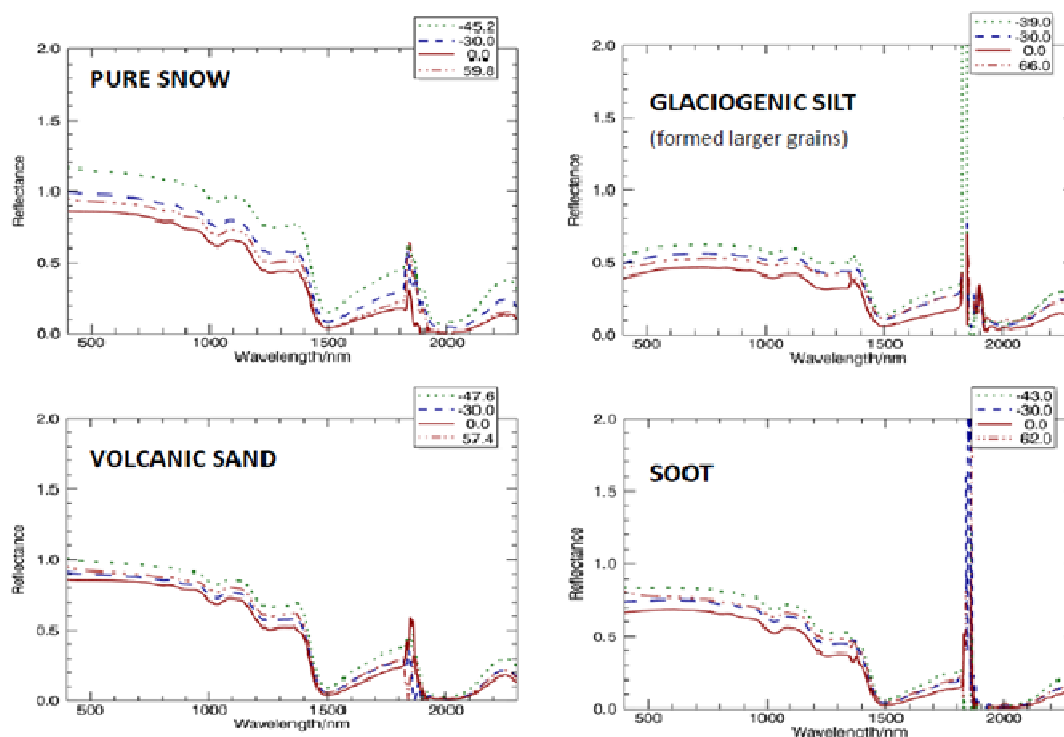


Figure 5.3 Spectral reflectance measurements of absorbing impurities on snow during the first days of the artificial deposition in April 2013 (reflectance spectrum in 4 directions).

by half (Figure 5.3). Two weeks after the deposition, the highest reflectance was measured at the reference spot and fine silt spot, whether the lowest reflectance came from the sooted and volcanic sand spots. The difference between the measurements in different part of the melting season was caused by fresh snow entering the spots. Overall, the larger volcanic sand particles reduced the reflectance more than the fine glaciogenic silt particles except that the small silt particles tended to form larger grains during several hours after deposition (Figure 5.4). As result, the newly formed larger grains lowered significantly the snow reflectance.



Figure 5.4 Melting, metamorphose and diffusion processes of the snow where glaciogenic silt was deposited. Left: Freshly deposited. Right: Several hours after dust deposition.

The impurities were observed to form larger particles and accelerate snow melt during the SoS-2013 campaign. It is agreed that this process of the "clumping mechanism" is likely to observe only during the artificial deposition of impurities on snow (Brandt et al. 2011). However, the Snow-Dust storm event that took place in Iceland in March 2013 confirmed that clumping mechanism can be observed in natural conditions. Figure 5.5 (right) shows detailed pictures of the formation of larger grains on snow in Reykjavik after the event. This dust travelled about 250 km before deposited on snow, but the layer of dust deposited on snow closer to the dust source was in order of several centimetres (Figure 5.5 left). We suggest that snow reflectance of such extreme deposition would be similar to reflectance of volcanic dust made in laboratory (see Chapter 5.3.3).

Darker snow surface after dust deposition lowered snow albedo, increased melt, and had direct effects on snow density. Added light-absorbing impurities were found to decrease the density of seasonally melting snow during the SOS-2013. Clean snow and snow with oil burner BC had higher snow density than snow where volcanic sand or wood burning BC was added. An independent laboratory experiment also showed that sooted snow released melt water sooner than unsooted.



Figure 5.5 The Icelandic Snow-Dust storm on March 6 2013, in Kirkjubaejarklaustur (left), caused an extreme volcanic dust deposition on snow. The impurities on snow in Reykjavik, 250 km from the dust source (right) were observed to form larger particles ("clumping mechanism") and accelerate snow melt. Left photo - courtesy of Ingveldur Gudny Sveinsdottir from Kirkjubaejarklaustur.

Our results show that dust deposited on snow reduces the snow reflectance similarly or substantially more than BC. Hadley and Kirchstetter (2012) found 20% decrease in snow albedo (at $\lambda = 0.412\mu\text{m}$) for the soot concentration $c = 1.68 \mu\text{g g}^{-1}$. However, Painter et al. (2007) observed the most pronounced decrease towards UV portion of the spectrum due to dust deposition. The snow reflectance at 412 nm was reduced by 50% when dust with $c = 0.37 \text{ mg g}^{-1}$ was deposited on a snow layer. This is due to the fact that the imaginary part of the dust refractive index increases towards UV, which is not the case for soot (Kokhanovsky, 2013). Painter et al. (2012) found that the positive radiative forcing of dust deposited on snow can be up to 75 W m^{-2} , while the snow cover duration is reduced by 51 days. The volcanic dust deposition on the Icelandic glaciers is estimated as $400 \text{ g m}^{-2} \text{ yr}^{-1}$ (Chapter 5.3.1), but several severe dust events bring extreme amounts of dust to the glaciers and snow surfaces in Iceland each year. Our results confirm that volcanic dust has strongly negative impacts on the snow and glaciers (Figure 5.5). Further snow campaigns or long term measurements of snow properties *in situ* are needed in Iceland.

5.3.3 Optical properties of volcanic dust and climate implications

Dust samples were also dried and investigated in laboratory. Both methods showed similar results. The reflectance of pure volcanic sand was about 0.03 measured by the spectroradiometer coupled with a contact probe and reflectance measured with FIGIFIGO ranged between 0.02 and 0.04. These are measurements nearly of "black body" equivalent. Glaciogenic silt had a reflectance ranging 0.05 and 0.19 (FIGIFIGO) caused by its light-brown colour in a dry state, but showing also a very low number. Clark et al. (1990) compared 33 dust samples and the spectral reflectance was higher than our results for all of

them. Present laboratory experiments give first information on optical properties of volcanic dust from Iceland, which can be applied for suspended particles travelling in the atmosphere.

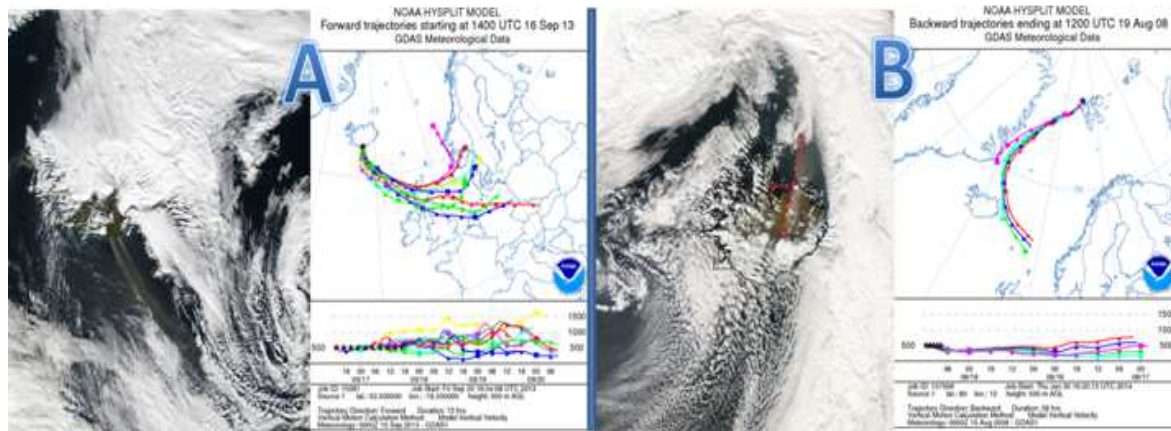


Figure 5.6 Aqua MODIS satellite images of dust storms in South (A) and Northeast (B) Iceland. Calculated forward (A) and backward (B) trajectories (HYSPLIT) for the events on September 16, 2013 reaching Europe (A), and August 17-18, 2008 reaching Svalbard (B).

There are strong indications that Icelandic dust particles in dust plumes travel long distances: i) high number of satellite images confirming dust plumes > 500 km in visible range of spectra (example Figure 5.6), ii) dust particles from Iceland were identified in Ireland, 1300 km distance (Ovadnevaite et al., 2009), iii) Icelandic dust particles were identified in the ice-core samples in central Greenland (Drab et al., 2002), and iv) dust periods retrieved from the ice-cores data during GISP2 project in Greenland (Donarummo et al., 2002) correlate with the NE Iceland dust frequency 1950-1990. However, there is a lack of case studies identifying the long range transport of Icelandic dust. Frequently used tool, the Positive Matrix Factorization (Paatero and Tapper, 1994), to detect the origin of atmospheric particulate matter, does not include any parametrization for Icelandic or volcanic dust yet. Many studies, especially from the Arctic region, relate identified volcanic dust particles to on-going eruptions.

Investigated physical properties of suspended volcanic dust in Iceland also indicate the predisposition of the dust to suspension: i) glaciogenic dust contains substantial numbers of close-to-ultrafine particles, which are sharp-tipped, curved, concave, and which contain large bubbles, ii) large volcanic particles from Iceland (up to 50 μm) travelled > 2000 km (Navratil et al., 2013), and iii) the amount of suspended dust during the severe dust storms is extremely high (Chapter 4). Further, Iceland is a region with high wind velocities and Icelandic cyclones are associated with the uplift of the surface air to the upper troposphere, while the tropopause is located lower in altitude than at more southern latitudes, at about 8 km height. This suggests that Icelandic aerosol can be transported into the stratosphere as well (Rose et al., 2006; Roesli, 2008), where the residence time is prolonged up to several weeks.

High frequency and severity of dust events in Iceland, optical and physical properties of volcanic dust particles, long-range transport of dust and dust deposition on snow, are together strong indicators that Icelandic dust has impacts on climate. We suggest that Iceland is not only a local source of dust, but could be a long-term contributor to Arctic and European air pollution. Optical and some physical properties of volcanic dust are more similar to BC, the most powerful absorbing aerosol, than crustal dust. Direct radiative forcing of mineral dust is concluded as negative in the IPCC report (IPCC, 2013), while indirect radiative forcing of dust deposited on snow is positive (Painter et al., 2012). Our results show that Icelandic volcanic dust is a strongly light-absorbing aerosol both directly and indirectly. Dust similarities in the properties and effects with the BC aerosol allow us to label the Icelandic volcanic dust as “naturally produced BC”, despite that volcanic sands are assumed not to contain BC (Dadic et al., 2013; Chapter 3, 5.3.2). We believe that our results may have potential in reducing the uncertainties (IPCC, 2013) related to the effect of dust on snow melt and climate change.

The Icelandic dust deposition is estimated to influence an area of $> 500,000 \text{ km}^2$ in total (Chapter 5.3.1). Such high-latitude dust events (> 9 dust events annually are in winter or at sub-zero temperatures) have the potential to contribute to Arctic warming. Icelandic glaciers are currently retreating due to climate change (Bjornsson and Palsson, 2008). Dust emissions are likely to increase over the next decades with retreating glaciers. Large floodplains will be subjected to intense aeolian forces, while a warmer climate will also enhance the radiative forcing effect over these dark surfaces.

6 Summary and conclusions

This thesis provides a new and comprehensive perspective of the dust aerosol production from natural sources in Iceland. It presents results from the pioneer field experiments of atmospheric and snow dust measurements as well as the long-term trends in frequency and severity of dust storm events in Iceland. Air quality in Iceland, which is generally considered good, is often impaired by dust suspension causing high particulate matter concentrations. Long-term dust frequency in Iceland is similar to major desert areas of the world (Mongolia, Iran, USA, China). Frequent volcanic eruptions and dust haze increase the number of dust events fourfold, resulting in 135 dust days annually. However, synoptic codes for volcanic ash resuspension and dust haze are not included in the conventional dust studies from the major deserts. Iceland is the most active polar desert in the Arctic/sub-Arctic region.

Dust deposition of 31 – 40 million tons affects areas of $> 500,000 \text{ km}^2$ annually while some dust plumes are spanning $> 1000 \text{ km}$ at times. Dust is also distributed over glaciers (about 4.5 million t annually) and oceans (6 – 14 million t annually). Dust deposited on glaciers and snow lowers the snow albedo and reduces the snow density. This leads to fast melting and snow cover reduction, possibly shortening snow melt to up to fifty days (Painter et al., 2012). The oceanic deposition of Icelandic dust rich in iron can potentially have marked influence on the primary productivity in oceans around Iceland, especially in spring and late summer.

Icelandic dust differs to crustal mineral dust, such as from the Sahara or Gobi deserts. The investigations of physical properties of Icelandic dust reveal major differences in mineralogy, geochemical compositions, shapes, sizes and colour, compared to crustal dust. Icelandic dust is of volcanic origin, dark in colour with sharp-tipped shards and large bubbles. About 75 – 80 % of the particulate matter is volcanic glass rich in heavy metals, such iron and titanium. Alkali glasses often have complicated pipe-vesicular structures more similar to the asbestos particles or Black Carbon than mineral dust, and may pose health risks. Suspended dust at the glacial dust source consists of extremely high number of close-to-ultrafine particles with the similar number concentrations as reported during the active eruptions. Glacial and water processes are steadily reworking the sediment, so that the very fine material can be easily suspended, even during moist conditions with low winds. However, giant particles such 50 – 100 micrometers can travel distances over 2000 km, likely as result of the unique morphology. Measurements of a severe dust storm after the Eyjafjallajökull eruption in 2010 captured grains $> 2 \text{ mm}$ and the aeolian transport exceeded amount of 11 tons of dust over one meter transect. This places Icelandic dust storms among the most extreme wind erosion events recorded on Earth.

The Sea Level Pressure oscillation controlled whether dust events occurred in NE (16.4 dust days annually) or in southern part of Iceland (about 18 dust days annually). The Arctic dust events (NE Iceland) were typically warm and during summer/autumn (May-

September) while the Sub-Arctic dust events (S Iceland) were mainly cold and during winter/spring (March-May). The most dust-frequent decade in S Iceland was the 1960s while the most frequent decade in NE Iceland was the 2000s. A total of 32 severe dust storms (visibility < 500 m) was observed in Iceland with the highest frequency during the 2000s in S Iceland. About half of dust events in S Iceland occurred in winter or at sub-zero temperatures. This confirms that Iceland is among the dustiest areas of the world and dust is emitted the year-round.

Optical properties of volcanic dust both on snow and in laboratory are found to be similar to Black Carbon, the most powerful absorbing aerosol. These first experiments indicate that climate forcing of Icelandic volcanic dust is different to that concluded on mineral dust in the IPCC report in 2013. Icelandic volcanic dust tends to act as a positive climate forcing agent both directly and indirectly.

The long-range transport and year-round activity of volcanic dust suggest that Icelandic dust have the potential to contribute to the Arctic warming. Iceland is not only a substantial source for local air pollution; it has likely long-term effects on the Arctic and European air pollution.

7 Future research

High-latitude sources of dust air pollution, such as Iceland, should be identified and classified in detail, considering that the air pollution is also affecting the Arctic and European region. Here are presented findings from two of seven major dust sources in Iceland, which differ in physical properties of dust aerosol. Information about the major direct source for the Arctic dust events, Dyngjúsandur, is missing. A network of environmental stations to measure the particulate mass and number concentrations around all these sources is necessary to quantify and verify the calculated amounts of dust air pollution emitted from Iceland. Ground based measurements should be expanded to the upper atmosphere to understand the vertical extent and aerosol stratification of dust pollution events.

Analyses of the geochemistry and mineralogy of different Icelandic dust types should be concluded in the Reference Material Database (RMD), which would serve as a platform giving continuous information for the Positive Matrix Factorization (PMF). The PMF is globally used by the scientific community to detect the origin of the atmospheric particulate matter. A special emphasis should be given to coarse-grained particles carried over long distances, which are often excluded from long-range transport studies and models. The RDM can be also applied for the future volcanic eruptions in Iceland.

New findings on dust frequency, mass and number concentrations and physical properties of Icelandic dust, are of interest to the medical community, to assess the impact of Icelandic dust on human health and air quality, with emphasis on the special characteristics of the dust (sharp, porous, high heavy metal concentration etc.). There is a need to link dust emission research and health records from various parts of Iceland, in order to estimate possible health effects. Furthermore, increased dust measurements are needed to develop health risk warning systems. High iron content in volcanic dust and large areas of oceanic dust deposition should be related to the research of marine ecosystem.

The first steps towards the parameterization for Icelandic volcanic dust are presented in this study. Future work should implement this information to the model. This would help us to understand main patterns in long-range transport of Icelandic dust and establish dust forecasting. Further, a network of aerosol stations in Greenland and Svalbard are “on alert” for the future dust events to obtain the evidence of the presence of Icelandic particles. We need more collaborators from Europe, where only the Central and South-Western European colleagues are prepared for such dust events.

It is very important to provide more measurements on optical properties of volcanic dust, both direct (via suspension, single particle studies) and indirect (snow and glacier deposition). Icelandic glaciers are currently retreating due to climate change. Dust emissions are likely to increase over the next decades with retreating glaciers as some of the major dust source areas leave behind larger floodplains subjected to intense aeolian

redistribution of fine sediments. It is important to increase understanding of the aeolian nature of these major dust source areas, including loading, erosion processes, deflation, and dust generation. The indirect effects of volcanic dust deposition on snow and glaciers should be investigated *in situ* in Iceland as a part of a COST-SNOW ES1404 action. We believe that the results presented here and future campaigns and experiments have a strong potential in reducing the uncertainties related to the effect of suspended dust on climate and effects of dust on snow melt and climate change in the future IPCC predictions. It should be emphasized that the volcanic dust has likely opposite climate effects to the crustal mineral dust, and it has significance in the large context of Arctic warming. Further, the incandescent temperature of volcanic dust should be studied in comparison to Black Carbon, the most powerful climate forcing aerosol.

We expect to conduct regular balloons flights of the small aerosol counter LOAC, in particular, during dust events, to determine the vertical extend of the plume and the nature of the suspended solid particles in near future. Further, a ground-based version of LOAC could be installed in Iceland, in order to determine the ambient air pollution and to determine accurately the size distribution of the particles.

Our ambitions for the future are to establish a multidisciplinary research network and advisory system for high-latitude dust monitoring and air pollution. This network shall also include short-term inputs of natural particulates to the atmosphere, such as volcanic eruptions, or cases of extra-terrestrial dust entering the Earth's atmosphere.

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