Volcanic sulphate and Arctic dust plumes over the North Atlantic Ocean

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Abstract. High time resolution aerosol mass spectrometry measurements were conducted during a field campaign at Mace Head Research Station, Ireland, in June 2007. Observations on one particular day of the campaign clearly indicated advection of aerosol from volcanoes and desert plains in Iceland which could be traced with NOAA Hysplit air mass back trajectories and satellite images. In conjunction with this event, elevated levels of sulphate and light absorbing particles were encountered at Mace Head. While sulphate concentration was continuously increasing, nitrate levels remained low indicating no significant contribution from anthropogenic pollutants. Sulphate concentration increased about 3.8 μg/m^3 in comparison with the background conditions. Corresponding sulphur flux from volcanic emissions was estimated to about 0.3 TgS/yr, suggesting that a large amount of sulphur released from Icelandic volcanoes may be

distributed over distances larger than 1000 km. Overall, our results corroborate that transport of volcanogenic sulphate and dust particles can significantly change the chemical composition, size distribution, and optical properties of aerosol over the North Atlantic Ocean and should be considered accordingly by regional climate models.

Key Words: volcanic sulphate aerosol, Icelandic dust, particle acidity, aerosol properties

1. Introduction

Sulphuric acid and sulphate aerosols are essential precursors for cloud formation over the oceans (Charlson et al., 1987). They act as a cloud condensation nuclei and affect the number of cloud droplets increasing cloud albedo and may also lead to changes in precipitation (Textor et al., 2004). Nss-sulphate over the oceans can originate from both natural and anthropogenic sources and concentrations vary widely as a function of time and location (Savoie et al., 2002). Volcanic emissions of SO_4 and its sulphur precursor gases can significantly contribute to the natural SO_4 budget, even at the quiescent stage (Berresheim and Jaeschke, 1983; Glasow et al., 2008). According to Graf et al. (1997) the non-eruptive volcanic degassing of SO_2 and subsequent sulphate formation is estimated to be responsible for 24% of the global annual direct radiative forcing of sulphate aerosols. The annual SO_2 flux into the atmosphere from Icelandic volcanoes is about 0.8-1.0 Tg/yr (Halmer et al., 2002) thus representing one of the largest atmospheric sulphur sources in the North Atlantic region. These emissions may substantially contribute to new particle formation over this region and modify the composition of air masses, for example, of clean polar origin (O'Dowd and Smith, 1993).

Furthermore, Iceland has extensive arid regions, which are seldom reported in surveys, because the climatic environment and the composition of the sand make these deserts unusual. The sand

originates largely from glacial margins, glacio-fluvial and volcanic deposits, and sedimentary rocks (Arnalds et al., 2001). Strong winds can generate dust storms over these deserts, and dust plumes may be transported over great distances impacting air quality in the British Isles and continental Europe (NASA, 2008; Prospero et al., 2008). Although dust storms may at times be relatively rare events, Prospero et al. (2008) pointed out that glaciers in Iceland have been retreating in recent decades and that this trend is expected to continue with changing climate. It is difficult to track the sulphate and dust plumes by composition analysis which is typically done with off-line techniques in long-term monitoring programmes such as Global Atmosphere Watch (GAW). Here we report real-time observations using a quadrupole aerosol mass spectrometer (Q-AMS) at the Mace Head Atmospheric GAW Research Station, on the west coast of Ireland, which enabled high time resolution measurements of size spectra and concentrations of organic and inorganic species in the sampled particles. Together with other real time measurement techniques operated at this station Q-AMS registered volcanic sulphate and Icelandic dust plumes in air advected over the North East Atlantic Ocean. Concurrent observations of aerosol size evolution and impact on light scattering are also discussed.

2. Experimental

A four-week field campaign was conducted in June 2007 at Mace Head Research Station, Ireland (53°19′ N, 9° 54′ W). The site has been described in detail by Jennings et al. (2003) and O'Connor et al. (2008). It is located on a peninsula and the wind direction sector between 190° and 300° is from the open North Atlantic Ocean providing excellent conditions for carrying out marine aerosol measurements.

All aerosol instruments were placed in the shore laboratory about 100 m from the coastline and 5 m above mean sea level, MSL. They were connected to the laminar flow community air

sampling system, which is constructed from a 100 mm diameter stainless-steel pipe with the main inlet at 10 m above ground level.

The size resolved non-refractory chemical composition of submicron aerosol particles was measured with an Aerodyne Quadrupole Aerosol Mass Spectrometer (Q-AMS, Aerodyne, Billerica, MA). The instrument is described by Jayne et al. (2000). Ionization efficiency was calibrated twice (at the very beginning and the end of the campaign) by 300 nm dry ammonium nitrate particles, according to the method described by Jimenez et al. (2003) and Allan et al. (2003). Measurements were performed with a time resolution of 5 min, with a vaporizer temperature of about 600 °C. A collection efficiency of CE = 0.5 was applied for the measurement period discussed here. According to laboratory experiments (Matthew et al., 2008, Middlebrook and Bahreini, 2008) the AMS collection efficiency depends on the particle chemical composition and relative humidity of the environment. They showed that at relative humidity about 80% and neutralization of sulphate particles about 50-60 %, which was the case during the analysed event, collection efficiency of AMS is about 0.5.

The total aerosol scattering coefficient and the hemispheric backscattering coefficient were measured with a TSI (TSI Incorporated, Shoreview, MN) Model 3563 three-wavelength (450; 550; 700 nm) integrating nephelometer.

Aerosol absorption measurements were performed with the Thermo scientific (Thermo Fisher Scientific Inc., Waltham, MA) multi-angle absorption photometer (MAAP) instrument, model 5012. This instrument calculates absorbance from particles deposited on a filter using measurements of both transmittance and reflectance at two different angles.

Aerosol size distributions were measured by a scanning mobility particle sizer (SMPS). The SMPS system is composed of a differential mobility analyzer (DMA, TSI model 3071), a

96 condensation particle counter (CPC, TSI model 3010), an aerosol neutralizer (TSI 3077), a
97 control unit, and a data logging system.
98 A volatility-hygroscopic-tandem differential mobility analyser (VHTDMA) was employed to
99 simultaneously measure the volatile and hygroscopic properties of particles ranging from 10 nm
100 to 110 nm in (mobility) diameter (Johnson et al., 2004). Comparison of VHTDMA signatures
101 measured in the field with those of laboratory aerosols of known chemical composition were
102 used to infer the composition of the atmospheric particles and their mixing state. A thorough
103 description of the VHTDMA and its operating procedures has been provided by Fletcher et al.
104 (2007).
105 Radon (^{222}Rn) and lead (^{212}Pb) isotope concentrations were determined using the active deposit
106 method. The absolute error is estimated to be in the order of 20% (Polian et al., 1986; Biraud et
107 al., 2000).
108 Backward trajectories up to 96 hours were calculated, using the National Oceanic and
109 Atmospheric Administration (NOAA) hybrid single-particle lagrangian integrated trajectory
110 (HY-SPLIT) model ((Draxler and Hess, 1997). Trajectories were calculated for every hour at
111 three different altitudes: 1000, 500 and 20 meters above ground level (AGL).
112
113 **3. Results**
114 On June 26, 2007, a high pressure system over Greenland and a low situated over the North
115 Atlantic Ocean and Ireland produced a north westerly flow with 8 m/s average local wind speed
116 and quite low temperature – about 12-14 ºC during the day. The relative humidity varied
117 between 80% and 90 % and particle concentration was less than 700 particles cm^{-3}, typical for
118 marine background air at Mace Head. Local wind direction shifted from 360º to 270º and
119 between 11:00 UTC and 24:00 UTC it was from the typical marine sector (which extends

between 190º and 300º). The air mass trajectories gradually shifted from north to west, and from 04:00 UTC had passed over Iceland approximately three days earlier before reaching Mace Head (Fig. 1).

3.1. Evolution of the aerosol chemical composition

We observed a continuous increase in sulphate concentrations in advected air masses with trajectories crossing over Iceland. Over the period of 11 hours the non-sea salt (nss)-sulphate concentration increased from 1.2 to 4.6 (±0.9) $\mu g/m^3$ (Fig. 2). Concurrent nitrate levels remained low and largely unchanged indicating no major contribution from anthropogenic pollution.

A minor increase was observed in the level of organics, probably originating from phytoplankton blooms located south-west of Iceland (NASA, 2008). However, the lack of correlation (R^2=0.02) between organic and sulphate trends suggests that the observed increase in sulphate concentration was associated with a different source. Moreover, marine biogenic nss-sulphate concentrations at Mace Head station as reported by Yoon et al. (2007) are less than 0.8 $\mu g/m^3$. In addition, the concurrent increase in radon concentrations (Fig. 2) supports a predominantly land origin of sulphate in this air mass. Radon concentrations were initially elevated due to regional contributions from Ireland but then decreased when trajectories shifted from north to north-west. Later (from 12:00 UTC) it started increasing again in conjunction with air mass passage over Iceland. The radon temporal trend followed the sulphate trend. Generally, the radon concentration on June 26 was lower than 400 mBq/m^3 indicating an oceanic air mass (Messager et al., 2008) and little to no contact with land over the past 2-3 days. This is consistent with the measurements of ^{212}Pb as lead concentration decreased after 12:00 UTC and stayed at low level until the end of the day. ^{222}Rn and ^{212}Pb are both emitted by soils and can be used as continental tracers. However, they have different radioactive decay time: 3.8 days for ^{222}Rn and 10.6 hours

144 for ^{212}Pb. Therefore, a decrease in concentration of the latter tracer showed the absence of a
145 recent contact with land, whereas increase in the radon concentration indicated a remote land
146 influence.
147 From these results we assume that the observed increase in sulphate concentration (3.4 μg/m^3
148 above background level) was entirely caused by advection of volcanic sulphur emissions from
149 Iceland. Using the time period of 11 hours and the 3.4 μg/m^3 increase in nss-sulphate levels we
150 estimated the total sulphur flux from Icelandic volcanoes assuming that all SO_2 had been
151 converted to sulphate. The measured increase in concentration was multiplied by the total
152 volume of the plume which could be derived using the FLEXPART model (Stohl et al., 2005).
153 The mixed layer height was assumed to be 1 km. We calculated a sulphur flux of 820 t S/d.
154 Assuming constant emissions this result was scaled to the yearly sulphur flux to compare it with
155 previous estimates of annual fluxes of sulphur from Icelandic volcanoes and from
156 dimethylsulphide (DMS) emissions from the North Atlantic.
157
158 *3.2. Volcanic aerosol acidity*
159 The sulphate particles were only partially neutralized by ammonium based on the results
160 obtained by both the Q-AMS and the Volatility Hygroscopic Tandem Differential Mobility
161 Analyzer (VHTDMA). Q-AMS measured at least 35% of the total sulphate mass being a pure
162 sulphuric acid (Fig. 3) while according to VHTDMA, H_2SO_4 constituted about 26% of total
163 sulphate volume in 50nm particles.
164 The modified marine air flow touching the west coast of Ireland (Fig. 1a) in conjunction with
165 northerly wind direction brought nearly neutralized sulphate particles in the form of ammonium
166 sulphate and bisulphate (00:00 UTC - 01:00 UTC, Fig. 3). However, with trajectories shifting to
167 the west and air masses coming from the marine sector (Fig. 1b, c), the degree of neutralization

168 decreased and sulphuric acid constituted about 50 % of the total sulphate mass (10:00 UTC-
169 23:00 UTC, Fig.3).
170 Figure 4 displays a VHTDMA scan of 50 nm particles completed between 13:55 UTC and 15:26
171 UTC on June 26. The volatility curve (particle volume fraction remaining (V/Vo) against
172 thermo denuder temperature) had 2 distinct volatilisation steps. The first component evaporated
173 at 80-140ºC and the second component evaporated at 160-220ºC. Comparison with a VHTDMA
174 scan of laboratory generated, partially neutralised sulphuric acid (H_2SO_4) particles (Johnson et al.
175 2005) suggests that the more volatile component is H_2SO_4 and the less volatile component is
176 partially neutralised H_2SO_4 (ammoniated sulphate). For the ambient scan the volume fraction of
177 H_2SO_4 is 26%. A repeat scan conducted between 16:15 UTC and 16:53 UTC was very similar to
178 the original scan. The volume fraction of H_2SO_4 was, again, 26%.
179 The degree of neutralisation was calculated assuming that any ammoniated sulphate existed as
180 letovicite ($(NH_4)_3H(SO_4)_2$). This assumption is based on the measured fraction of sulphuric acid
181 and the H_2O-$(NH_4)_2SO_4$-H_2SO_4 phase diagram presented by Tang et al. (1978). The per-particle
182 volumes of H_2SO_4 and $(NH_4)_3H(SO_4)_2$ were calculated from the initial particle diameters and
183 particle diameters at 150ºC and 220ºC. These volumes were converted to mole numbers using
184 density and molecular weight. The density of H_2SO_4 was calculated according to the
185 parameterisation of Tang (1996). Finally, considering the stoichiometry of letovicite the
186 ammonium to sulphate ratio was calculated to be 110.5%.
187 However, with respect to particle size the Q-AMS covered the 40-1000 nm diameter range of
188 particles, whereas VHTDMA measured the composition of only 50 nm particles at that time.
189 Therefore the combined results suggest that there was a greater degree of neutralisation for
190 smaller particles. Previous studies e.g., by Fletcher et al. (2007) at Cape Grim station and
191 Tomlinson et al. (2007) over the south-eastern Pacific also reported a relatively stronger

192 neutralization in lower particle sizes. They suggested that size dependent acidity could be
193 explained by H_2SO_4 being added to the aerosol by in-cloud aqueous phase processes faster than
194 it can be neutralized by the limited available ammonium. On the other hand, smaller particles
195 may simply be neutralized by the limited ammonium more readily due to their higher surface-
196 volume ratio (McMurry et al. 1983). In our study both instruments showed the presence of pure
197 sulphuric acid in the particles which is consistent with previous study of Satsumabayashi et al.
198 (2004) who showed that aerosols affected by volcanic emissions were strongly acidic as the
199 excess amount of SO_4^{2-} exhausted ammonium and partially expelled NO^{3-} and Cl^-. Furthermore,
200 Stothers and Rampino (1983) demonstrated that the largest acidity signals in old Greenland ice
201 are due to European (Mediterranean and Icelandic) volcanic eruptions. Typically, the ammonium
202 to sulphate ratio measured near a volcanic source is much lower compared to a site further
203 downwind (Johnson and Parnell, 1986; Mather et al., 2003).
204
205 *3.3. Aerosol radiative absorption and scattering*
206 In addition to sulphate plume, an increase in absorption was registered by multi-angle absorption
207 photometer (MAAP). Figure 6 shows that noticeable amount of absorbing material was
208 measured between 16:15 UTC and 20:30 UTC. The Moderate Resolution Imaging
209 Spectroradiometer (MODIS) flying on NASA's Terra satellite captured the image of dust plumes
210 blowing off the southern coast of Iceland over the North Atlantic Ocean on June 23, 2007 (Fig.
211 5). Thus following this event, elevated concentrations of light absorbing particles were
212 encountered at Mace Head (Fig. 6). MAAP is a filter-based technique for measuring aerosol light
213 absorption, it rely on the change in transmission of light through a filter as it is loaded with
214 aerosol to determine the aerosol absorption coefficient. Similar to other filter-based techniques
215 measurements can be biased during specific conditions (Lack et al., 2008) such as high loadings

of dust aerosol. Dust particles can both absorb and scatter sunlight (Miller et al., 2004). Consistent with these properties our measurements showed a good correlation between light absorption and both back and total (back plus forward) scattering coefficients (Fig. 6). Several previous studies demonstrated that dust events increased scattering coefficients and nephelometer could be used as a good indicator of dust particle advection (Ichoku C. et al. 1999; Kim et al., 2004; Derimian et al., 2006). During advection of Icelandic dust to Mace Head , the total scattering coefficient followed similar trends at all three wavelengths (450, 550, 700 nm) whereas the backscattering coefficient was more spread between different wavelengths of light although they showed a similar time trend. These differences between wavelengths became significant (~15-20%) when the fraction of the organic aerosol material increased (12:00-13:00 UTC, 20:00-21:00 UTC) showing an inhomogeneous particle size distribution. In addition, Figure 6 shows that the light-scattering associated by sulphate aerosol was lower compared to the dust particles. Aerosol light scattering coefficients peaked at the same time as absorption maximum and only qualitatively followed sulphate concentration increase.

3.4. Aerosol size distribution

During the studied event, the aerosol size distribution was bimodal with well defined Aitken and accumulation modes. Figure 7 presents the temporal evolution of the aerosol size spectrum. Initially, a higher fraction of sulphate resulted in the growth of both accumulation and Aitken mode particles. However, this changed later due to the increasing fraction of dust particles. Consequently from 16:30 UTC we observed both higher absorption levels and a concurrent change in the size distribution (17:30 UTC). The Aitken mode diameter continued to increase while the accumulation mode diameter shifted towards smaller sizes (Table 1). In addition, dust particles increased the number concentration in both the Aitken and accumulation modes.

From 20:30 UTC air masses were advected from the North Atlantic Ocean which had not passed over Iceland. In this context, a rapid decrease in sulphate concentrations and significant change in the aerosol size spectrum was observed resulting in typical aerosol size distribution for a very clean air mass at Mace Head (Yoon et al., 2007).

4. Discussions

In general, the climate relevant implications of volcanic eruptions have been recognized in previous studies (Toon and Pollack, 1980; Gilliland, 1982; Rampino and Self, 1984; Minnis et al., 1993; Robock A., 2000). However, quasi-continuous non-eruptive emissions may have an even stronger incremental climatic impact than the relatively brief cataclysmic eruptions (Graf et al., 1997). For example, the annual sulphur input into the atmosphere from Icelandic volcanoes is of the same range as the total sulphur flux from the dimethylsulphide over the whole North Atlantic Ocean, 0.5 TgS/yr (Halmer et al., 2002) and 1.3 TgS/yr (Kettle and Andreae, 2000) respectively. Moreover, we estimated that Icelandic volcanogenic sulphur flux was about 0.3 TgS/yr (see previous section for details) suggesting that a large amount of sulphur emissions from Icelandic volcanoes can reach distances larger than 1000 km (Iceland to Ireland distance is approximately 1300 km). Such calculations, based on one event, should be considered as a rough estimate of the regional impact of degassing sulphur emissions; however it corresponds well with the previous calculations of total sulphur flux from Icelandic volcanoes (Halmer et al., 2002). Thus despite its relative point source character the distribution of sulphur from Icelandic volcanoes over the North Atlantic region was comparable to the larger scale input from marine biogenic sources.

The most obvious and well known volcanic effect is on solar radiation, since the sulphate particles are about the same size as the visible light wavelength (Robock, 2000). Though only a

264 relatively small fraction of sulphate is directly emitted from volcanoes, the bulk of the sulphate
265 aerosol is formed during the long range transport of the emissions by both gas and aqueous phase
266 oxidation of SO_2 (Hegg, 1985), affecting the resulting aerosol size distribution. Our observations
267 of a typical aerosol bimodal size distribution during the volcanic plume event suggest a
268 significant cloud processing of the aerosol along the trajectory path where accumulation mode
269 was formed by cloud activation of Aitken mode particles (Hoppel et al., 1986; Fitzgerald, 1991;
270 Bott, 1999; O'Dowd et al., 2000).
271 Presumably two processes took place during this transport: a new sulphate aerosol formation
272 and its subsequent neutralization by cations such as ammonium. As measurements near
273 volcanoes show highly acidic aerosol (Johnson and Parnell, 1986; Mather et al., 2003;
274 Satsumabayashi et al., 2004), the partially neutralised aerosol measured at Mace Head indicates
275 the chemical aerosol evolution during air mass transport. Along with the chemical transformation
276 the physical aerosol properties have been changing as well, with the increasing fraction of
277 sulphate resulting in the growth of both accumulation and Aitken mode particles. Moreover,
278 concurrent advection of dust particles from Icelandic deserts contributed to further modifications
279 of the aerosol properties. Dust particles were identified by two independent albeit indirect
280 measurements using the three-wavelength integrating nephelometer and the multi-angle
281 absorption photometer. We observed a corresponding increase by about 60% in light absorption
282 and scattering caused by advected sulphate and dust particles which indicates their strong
283 climatic implications.
284
285 **5. Conclusions**
286 Advection of volcanogenic sulphate aerosol from Iceland significantly increased sulphate
287 concentrations at Mace Head. Moreover it was shown that the corresponding volcanic plume

caused a substantial modification in the aerosol chemical composition and size distribution over the North Atlantic region during transport. Over the period of 11 hours the non-sea salt sulphate concentration increased by a factor of four while concurrent nitrate and organic levels remained low and largely unchanged. The sulphate particles were only partially neutralized. The Q-AMS measurements showed that at least 35% of the total sulphate mass was pure sulphuric acid in submicrometer (PM1) aerosol. Concurrent VHTDMA measurements showed that H_2SO_4 constituted about 26% of the total sulphate volume in particles of 50 nm diameter size. The combined results suggest that there was a relatively larger degree of neutralisation by ammonium in smaller particles. The predominant fraction of sulphate aerosol is assumed to have been formed by aqueous phase oxidation of SO_2 based on the efficiency of this process and the cloud cover associated with the air mass trajectories. A concurrent dust outbreak from Iceland increased the levels of absorbing material and the light-scattering. Aerosol light scattering coefficients peaked at the same time as absorption maximum and qualitatively followed sulphate concentration increase. Evaluated sulphur flux demonstrated that a large amount of the total (non-eruptive) sulphur emissions from Icelandic volcanoes can reach distances larger than 1000 km. These results in conjunction with previous research suggest that volcanogenic emissions and Aeolian dust from Arctic deserts in Iceland can be potentially significant regional sources of aerosols over the North Atlantic and therefore should be adequately considered in regional and global climate models.

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313

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Table1

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Time, UTC	11:30	14:00	15:50	17:30	19:00	20:00	21:30
Aitken mode median diameter, nm	34 ±0.1	34 ±0.1	35 ±0.1	41 ±0.2	46 ±0.3	52 ±0.3	36 ±0.2
Number concentration, $1/cm^3$	613±11	471±9	439±9	383±7	473±8	437±9	218±3
Accumulation mode median diameter, nm	162 ±4	184 ±3	191 ±4	186 ±2	163 ±4	154 ±2	209 ±4
Number concentration, $1/cm^3$	130±1	127±2	129±2	158±2	209±2	263±4	108±1

Captions

Fig. 1. Evolution of air mass back trajectories, calculated for June 26, 2007 (NOAA HYSPLIT): a) 02:00 UTC; b) 11:00 UTC; c) 19:00 UTC. Backward trajectories length was up to 96 hours at three different altitudes: 1000, 500 and 20 meters above ground level.

Fig. 2. Temporal trends of chemical composition of PM1 aerosol, measured by Q-AMS and concurrent radon concentrations on June 26, 2007.

Fig. 3. Time trends of ammonium to sulphate molar ratios in PM1 aerosol, measured by Q-AMS on June 26, 2007. There are 2 moles of ammonium for every mole of sulphate consequently when sulphate is fully neutralized in the form of ammonium sulphate the ratio is 200%, when sulphate is ammonium bisulphate ratio is 100%, and for particulate sulphuric acid ratio is 0.

Fig. 4. VHTDMA scan of 50 nm particles completed between 13:55 UTC and 15:26 UTC on June 26. Volume fraction remaining (V/Vo) and Hygroscopic Growth Factor (HGF) at 90% RH are both displayed as a function of thermo denuder temperature. Also plotted are the corresponding curves for 100nm partially neutralised H_2SO_4 particles measured in the laboratory by Johnson et al. (2005). The two steps in the laboratory measured volatility curve correspond to evaporation of H_2SO_4 (60-150ºC) and ammoniated sulphate (160-220ºC).

Fig. 5. Dust storm blowing off the southern coast of Iceland over the North Atlantic Ocean on June 23, 2007 (NASA, 2008).

Fig. 6. Enhancement of scattering coefficients and absorption due to increase in sulphate concentration and dust particles levels on June 26, 2007.

Fig. 7. Evolution of bimodal size distribution, measured by SMPS on June 26, 2007.

Table 1. Summary of the modal parameters obtained by fitting lognormal functions to aerosol number size distribution measured by SMPS in Mace Head on June 26.

Figure 1

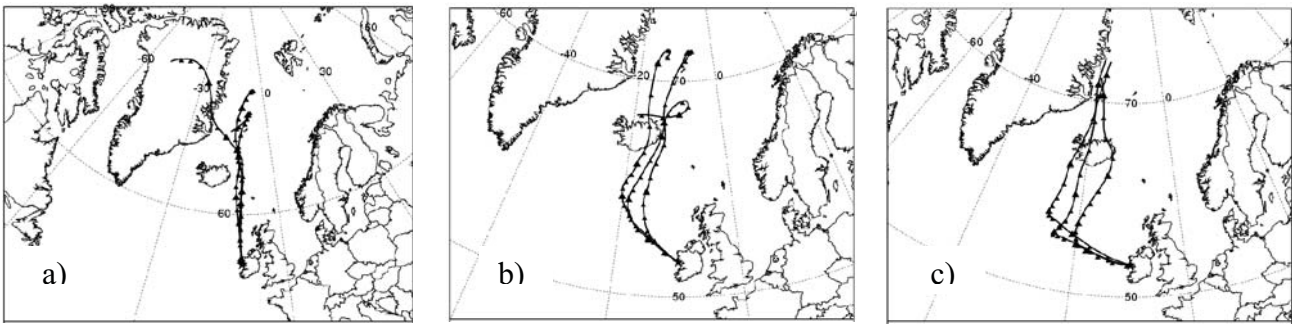


Figure2

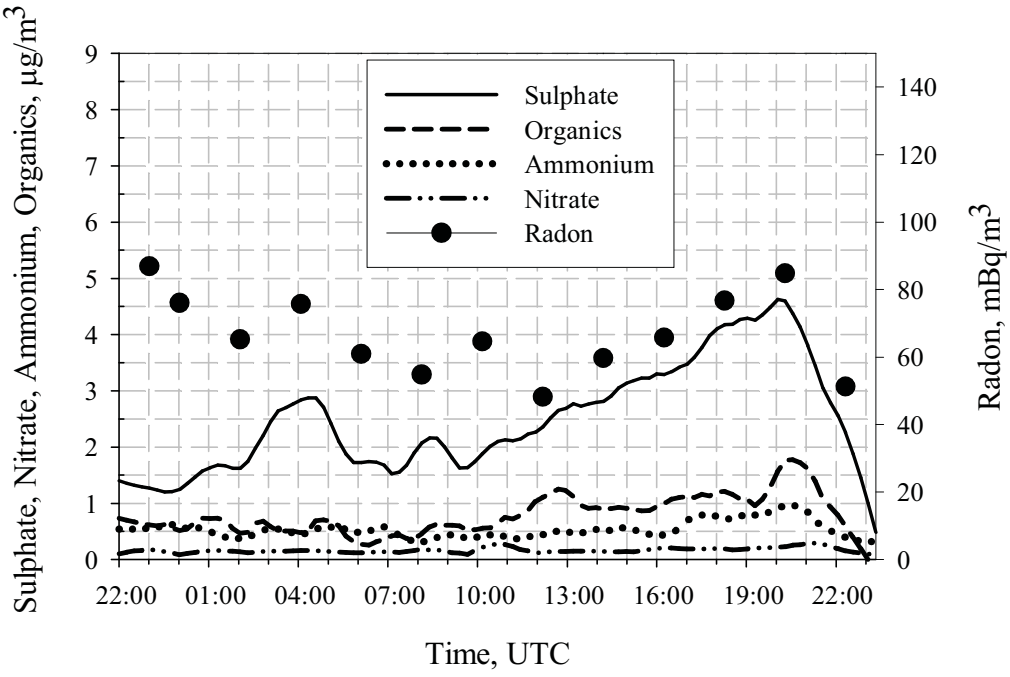

Sulphate
Organics
Ammonium
Nitrate
Radon
Sulphate, Nitrate, Ammonium, Organics, µg/m3
Radon, mBq/m3
0
1
2
3
4
5
6
7
8
9
0
20
40
60
80
100
120
140
22:00
01:00
04:00
07:00
10:00
13:00
16:00
19:00
22:00
Time, UTC

Figure 3

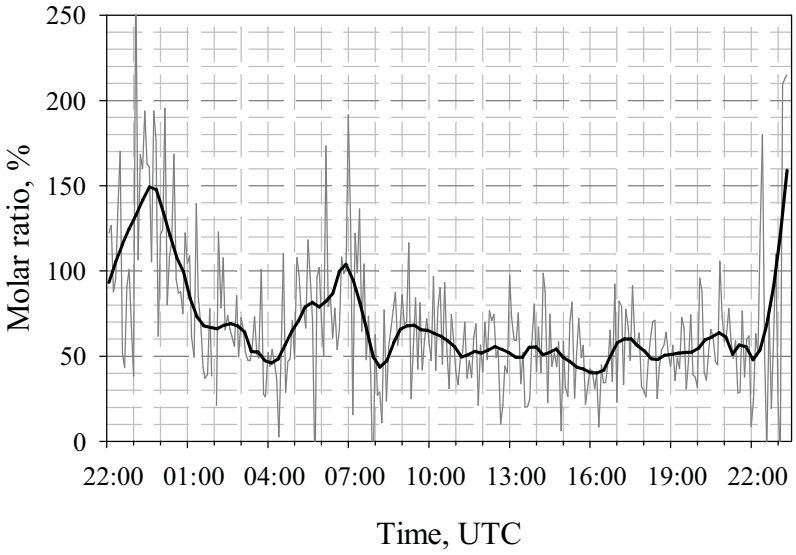

250
200
150
100
50
0
Molar ratio, %
22:00
01:00
04:00
07:00
10:00
13:00
16:00
19:00
22:00
Time, UTC

Figure 4

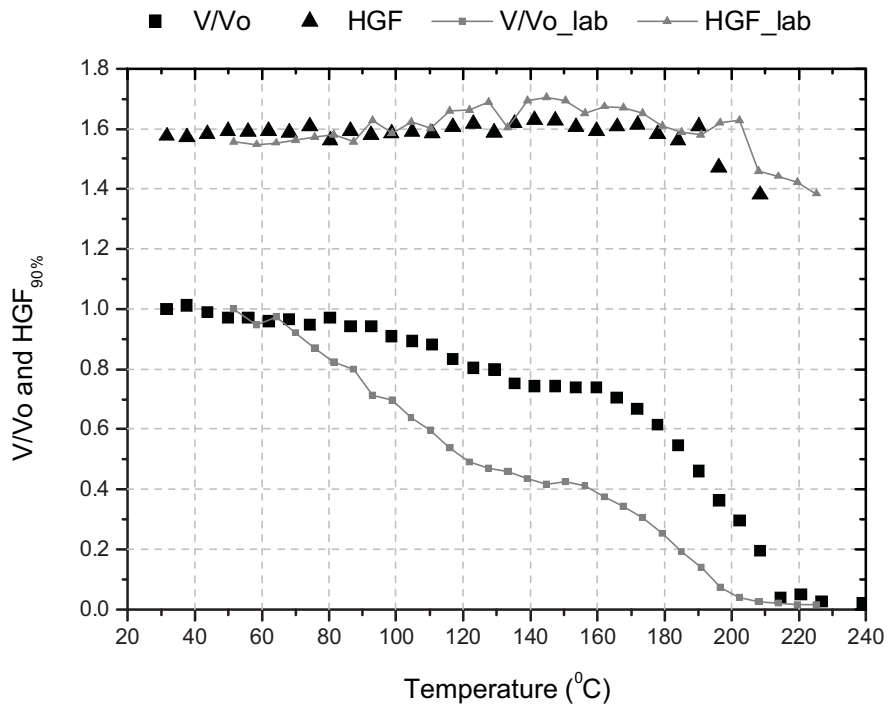

V/Vo
HGF
V/Vo_lab
HGF_lab
V/Vo and HGF90%
Temperature (0C)
1.8
1.6
1.4
1.2
1.0
0.8
0.6
0.4
0.2
0.0
20
40
60
80
100
120
140
160
180
200
220
240

Figure 5
Click here to download high resolution image

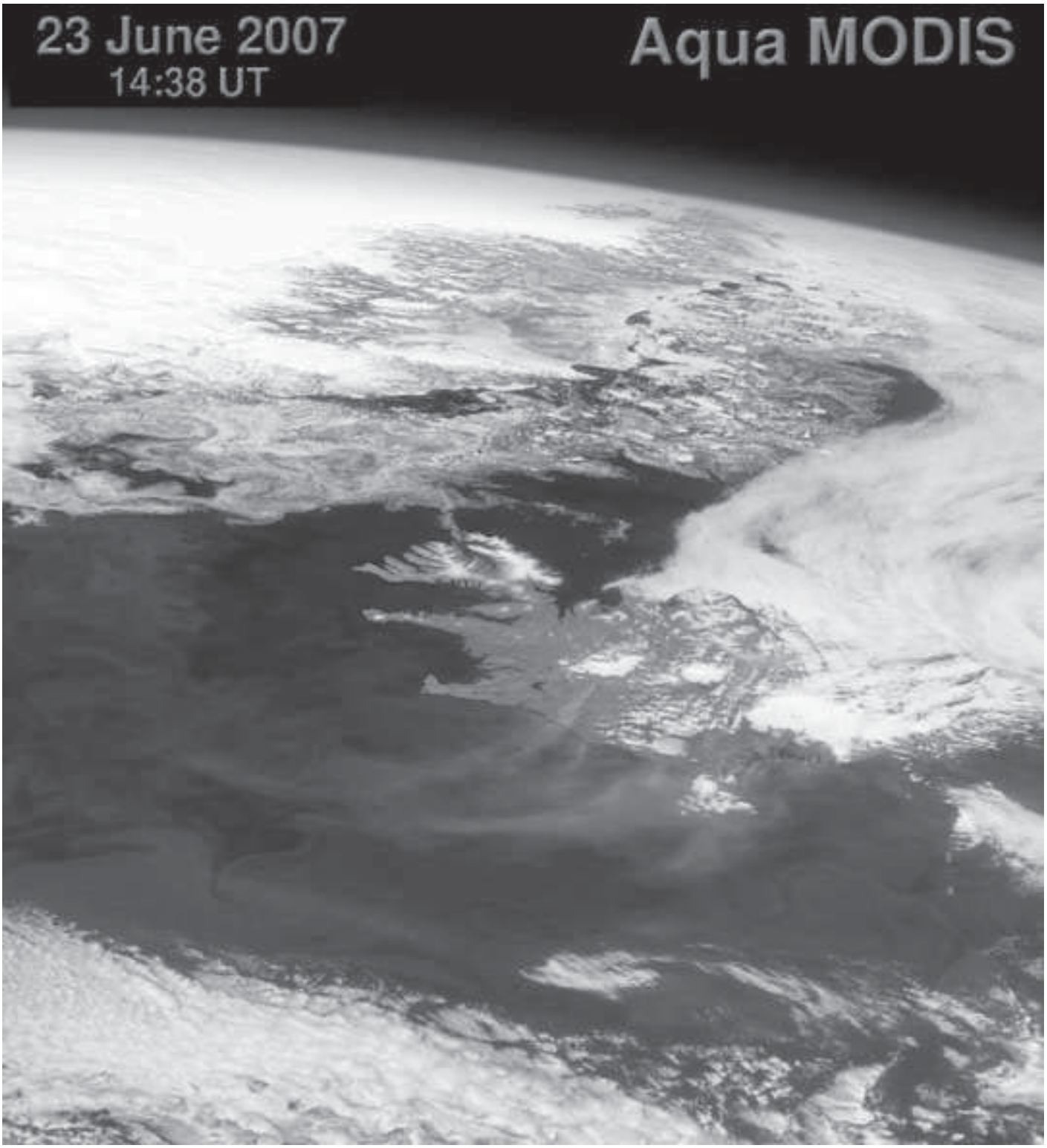


Figure 5 colour for the Web
Click here to download high resolution image

Figure6

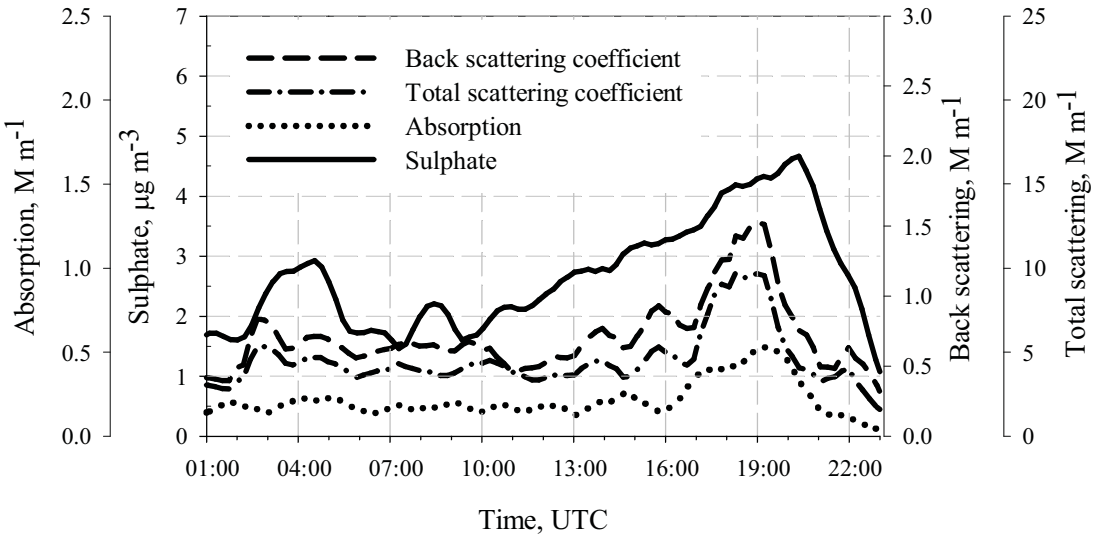
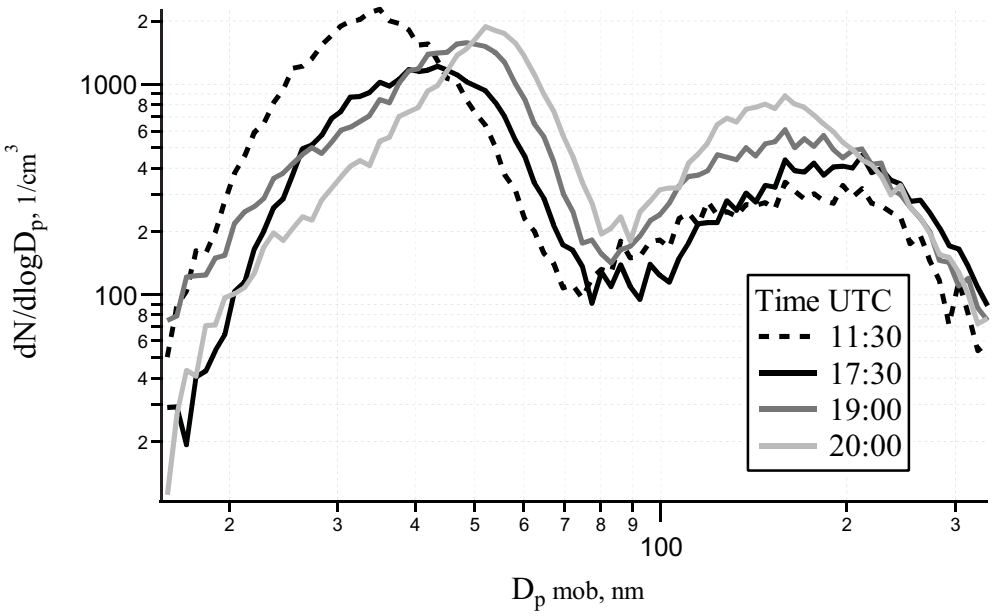

Absorption, M m-1
Sulphate, µg m-3
Back scattering, M m-1
Total scattering, M m-1
Back scattering coefficient
Total scattering coefficient
Absorption
Sulphate
01:00
04:00
07:00
10:00
13:00
16:00
19:00
22:00
Time, UTC

Figure 7


dN/dlogD$_p$, 1/cm^3
1000
100
2
3
4
5
6
7
8
9
100
2
3
D$_p$ mob, nm
Time UTC
11:30
17:30
19:00
20:00