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1   **Volcanic sulphate and Arctic dust plumes over the North Atlantic Ocean**

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3   J. Ovadnevaite[1, 2, *], D. Ceburnis[1], K. Plauskaite-Sukiene[2], R. Modini[3], R. Dupuy[1], I. Rimselyte[2],

4   M. Ramonet[4], K. Kvietkus[2], Z. Ristovski[3], H. Berresheim[1] and C. D. O'Dowd[1]

5

6   [1] School of Physics and Centre for Climate and Air Pollution Studies, Environmental Change

7   Institute, National University of Ireland, Galway, Ireland

8   [2] Institute of Physics, Savanoriu 231, LT-02300 Vilnius, Lithuania

9   [3] ILAQH, Queensland University of Technology, 2 George Street, Brisbane 4000 QLD, Australia

10   [4] Laboratoire des Sciences du Climat et de l'Environment (LSCE), Gif sur Yvette, France

11   *Corresponding author: E-mail address: *jurgita.ovadnevaite@nuigalway.ie*, Tel.:+35391492496,

12   Fax. :+35391494584, Address: School of Physics and Centre for Climate and Air Pollution

13   Studies, Environmental Change Institute, National University of Ireland, Galway, Ireland

14

15   **Abstract.** High time resolution aerosol mass spectrometry measurements were conducted during

16   a field campaign at Mace Head Research Station, Ireland, in June 2007. Observations on one

17   particular day of the campaign clearly indicated advection of aerosol from volcanoes and desert

18   plains in Iceland which could be traced with NOAA Hysplit air mass back trajectories and

19   satellite images. In conjunction with this event, elevated levels of sulphate and light absorbing

20   particles were encountered at Mace Head. While sulphate concentration was continuously

21   increasing, nitrate levels remained low indicating no significant contribution from anthropogenic

22   pollutants. Sulphate concentration increased about 3.8 $\mu g/m^3$ in comparison with the background

23   conditions. Corresponding sulphur flux from volcanic emissions was estimated to about 0.3

24   TgS/yr, suggesting that a large amount of sulphur released from Icelandic volcanoes may be

25 distributed over distances larger than 1000 km. Overall, our results corroborate that transport of

26 volcanogenic sulphate and dust particles can significantly change the chemical composition, size

27 distribution, and optical properties of aerosol over the North Atlantic Ocean and should be

28 considered accordingly by regional climate models.

29

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

31

32 **1. Introduction**

33 Sulphuric acid and sulphate aerosols are essential precursors for cloud formation over the oceans

34 (Charlson et al., 1987). They act as a cloud condensation nuclei and affect the number of cloud

35 droplets increasing cloud albedo and may also lead to changes in precipitation (Textor et al.,

36 2004). Nss-sulphate over the oceans can originate from both natural and anthropogenic sources

37 and concentrations vary widely as a function of time and location (Savoie et al., 2002). Volcanic

38 emissions of SO_4 and its sulphur precursor gases can significantly contribute to the natural SO_4

39 budget, even at the quiescent stage (Berresheim and Jaeschke, 1983; Glasow et al., 2008).

40 According to Graf et al. (1997) the non-eruptive volcanic degassing of SO_2 and subsequent

41 sulphate formation is estimated to be responsible for 24% of the global annual direct radiative

42 forcing of sulphate aerosols. The annual SO_2 flux into the atmosphere from Icelandic volcanoes

43 is about 0.8-1.0 Tg/yr (Halmer et al., 2002) thus representing one of the largest atmospheric

44 sulphur sources in the North Atlantic region. These emissions may substantially contribute to

45 new particle formation over this region and modify the composition of air masses, for example,

46 of clean polar origin (O'Dowd and Smith, 1993).

47 Furthermore, Iceland has extensive arid regions, which are seldom reported in surveys, because

48 the climatic environment and the composition of the sand make these deserts unusual. The sand

49 originates largely from glacial margins, glacio-fluvial and volcanic deposits, and sedimentary

50 rocks (Arnalds et al., 2001). Strong winds can generate dust storms over these deserts, and dust

51 plumes may be transported over great distances impacting air quality in the British Isles and

52 continental Europe (NASA, 2008; Prospero et al., 2008). Although dust storms may at times be

53 relatively rare events, Prospero et al. (2008) pointed out that glaciers in Iceland have been

54 retreating in recent decades and that this trend is expected to continue with changing climate. It

55 is difficult to track the sulphate and dust plumes by composition analysis which is typically done

56 with off-line techniques in long-term monitoring programmes such as Global Atmosphere Watch

57 (GAW). Here we report real-time observations using a quadrupole aerosol mass spectrometer

58 (Q-AMS) at the Mace Head Atmospheric GAW Research Station, on the west coast of Ireland,

59 which enabled high time resolution measurements of size spectra and concentrations of organic

60 and inorganic species in the sampled particles. Together with other real time measurement

61 techniques operated at this station Q-AMS registered volcanic sulphate and Icelandic dust

62 plumes in air advected over the North East Atlantic Ocean. Concurrent observations of aerosol

63 size evolution and impact on light scattering are also discussed.

64

65 **2. Experimental**

66 A four-week field campaign was conducted in June 2007 at Mace Head Research Station, Ireland

67 (53°19 ′ N, 9° 54 ′ W). The site has been described in detail by Jennings et al. (2003) and

68 O'Connor et al. (2008). It is located on a peninsula and the wind direction sector between 190°

69 and 300° is from the open North Atlantic Ocean providing excellent conditions for carrying out

70 marine aerosol measurements.

71 All aerosol instruments were placed in the shore laboratory about 100 m from the coastline and 5

72 m above mean sea level, MSL. They were connected to the laminar flow community air

73 sampling system, which is constructed from a 100 mm diameter stainless-steel pipe with the

74 main inlet at 10 m above ground level.

75 The size resolved non-refractory chemical composition of submicron aerosol particles was

76 measured with an Aerodyne Quadrupole Aerosol Mass Spectrometer (Q-AMS, Aerodyne,

77 Billerica, MA). The instrument is described by Jayne et al. (2000). Ionization efficiency was

78 calibrated twice (at the very beginning and the end of the campaign) by 300 nm dry ammonium

79 nitrate particles, according to the method described by Jimenez et al. (2003) and Allan et al.

80 (2003). Measurements were performed with a time resolution of 5 min, with a vaporizer

81 temperature of about 600 °C. A collection efficiency of CE = 0.5 was applied for the

82 measurement period discussed here. According to laboratory experiments (Matthew et al., 2008,

83 Middlebrook and Bahreini, 2008) the AMS collection efficiency depends on the particle

84 chemical composition and relative humidity of the environment. They showed that at relative

85 humidity about 80% and neutralization of sulphate particles about 50-60 %, which was the case

86 during the analysed event, collection efficiency of AMS is about 0.5.

87 The total aerosol scattering coefficient and the hemispheric backscattering coefficient were

88 measured with a TSI (TSI Incorporated, Shoreview, MN) Model 3563 three-wavelength (450;

89 550; 700 nm) integrating nephelometer.

90 Aerosol absorption measurements were performed with the Thermo scientific (Thermo Fisher

91 Scientific Inc., Waltham, MA) multi-angle absorption photometer (MAAP) instrument, model

92 5012. This instrument calculates absorbance from particles deposited on a filter using

93 measurements of both transmittance and reflectance at two different angles.

94 Aerosol size distributions were measured by a scanning mobility particle sizer (SMPS). The

95 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

120 between 190º and 300º). The air mass trajectories gradually shifted from north to west, and from

121 04:00 UTC had passed over Iceland approximately three days earlier before reaching Mace Head

122 (Fig. 1).

123

124 *3.1. Evolution of the aerosol chemical composition*

125 We observed a continuous increase in sulphate concentrations in advected air masses with

126 trajectories crossing over Iceland. Over the period of 11 hours the non-sea salt (nss)-sulphate

127 concentration increased from 1.2 to 4.6 ($\pm$0.9) $\mu g/m^3$ (Fig. 2). Concurrent nitrate levels remained

128 low and largely unchanged indicating no major contribution from anthropogenic pollution.

129 A minor increase was observed in the level of organics, probably originating from phytoplankton

130 blooms located south-west of Iceland (NASA, 2008). However, the lack of correlation (R^2=0.02)

131 between organic and sulphate trends suggests that the observed increase in sulphate

132 concentration was associated with a different source. Moreover, marine biogenic nss-sulphate

133 concentrations at Mace Head station as reported by Yoon et al. (2007) are less than 0.8 $\mu g/m^3$. In

134 addition, the concurrent increase in radon concentrations (Fig. 2) supports a predominantly land

135 origin of sulphate in this air mass. Radon concentrations were initially elevated due to regional

136 contributions from Ireland but then decreased when trajectories shifted from north to north-west.

137 Later (from 12:00 UTC) it started increasing again in conjunction with air mass passage over

138 Iceland. The radon temporal trend followed the sulphate trend. Generally, the radon

139 concentration on June 26 was lower than 400 mBq/m^3 indicating an oceanic air mass (Messager

140 et al., 2008) and little to no contact with land over the past 2-3 days. This is consistent with the

141 measurements of ^{212}Pb as lead concentration decreased after 12:00 UTC and stayed at low level

142 until the end of the day. ^{222}Rn and ^{212}Pb are both emitted by soils and can be used as continental

143 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

216   of dust aerosol. Dust particles can both absorb and scatter sunlight (Miller et al., 2004).

217   Consistent with these properties our measurements showed a good correlation between light

218   absorption and both back and total (back plus forward) scattering coefficients (Fig. 6). Several

219   previous studies demonstrated that dust events increased scattering coefficients and

220   nephelometer could be used as a good indicator of dust particle advection (Ichoku C. et al. 1999;

221   Kim et al., 2004; Derimian et al., 2006). During advection of Icelandic dust to Mace Head , the

222   total scattering coefficient followed similar trends at all three wavelengths (450, 550, 700 nm)

223   whereas the backscattering coefficient was more spread between different wavelengths of light

224   although they showed a similar time trend. These differences between wavelengths became

225   significant (~15-20%) when the fraction of the organic aerosol material increased (12:00-13:00

226   UTC, 20:00-21:00 UTC) showing an inhomogeneous particle size distribution. In addition,

227   Figure 6 shows that the light-scattering associated by sulphate aerosol was lower compared to

228   the dust particles. Aerosol light scattering coefficients peaked at the same time as absorption

229   maximum and only qualitatively followed sulphate concentration increase.

230

231   *3.4. Aerosol size distribution*

232   During the studied event, the aerosol size distribution was bimodal with well defined Aitken and

233   accumulation modes. Figure 7 presents the temporal evolution of the aerosol size spectrum.

234   Initially, a higher fraction of sulphate resulted in the growth of both accumulation and Aitken

235   mode particles. However, this changed later due to the increasing fraction of dust particles.

236   Consequently from 16:30 UTC we observed both higher absorption levels and a concurrent

237   change in the size distribution (17:30 UTC). The Aitken mode diameter continued to increase

238   while the accumulation mode diameter shifted towards smaller sizes (Table 1). In addition, dust

239   particles increased the number concentration in both the Aitken and accumulation modes.

240 From 20:30 UTC air masses were advected from the North Atlantic Ocean which had not passed

241 over Iceland. In this context, a rapid decrease in sulphate concentrations and significant change

242 in the aerosol size spectrum was observed resulting in typical aerosol size distribution for a very

243 clean air mass at Mace Head (Yoon et al., 2007).

244

245 **4. Discussions**

246 In general, the climate relevant implications of volcanic eruptions have been recognized in

247 previous studies (Toon and Pollack, 1980; Gilliland, 1982; Rampino and Self, 1984; Minnis et

248 al., 1993; Robock A., 2000). However, quasi-continuous non-eruptive emissions may have an

249 even stronger incremental climatic impact than the relatively brief cataclysmic eruptions (Graf et

250 al., 1997). For example, the annual sulphur input into the atmosphere from Icelandic volcanoes is

251 of the same range as the total sulphur flux from the dimethylsulphide over the whole North

252 Atlantic Ocean, 0.5 TgS/yr (Halmer et al., 2002) and 1.3 TgS/yr (Kettle and Andreae, 2000)

253 respectively. Moreover, we estimated that Icelandic volcanogenic sulphur flux was about 0.3

254 TgS/yr (see previous section for details) suggesting that a large amount of sulphur emissions

255 from Icelandic volcanoes can reach distances larger than 1000 km (Iceland to Ireland distance is

256 approximately 1300 km). Such calculations, based on one event, should be considered as a rough

257 estimate of the regional impact of degassing sulphur emissions; however they are in a good

258 agreement with the previous calculations of total sulphur flux from Icelandic volcanoes (Halmer

259 et al., 2002). Thus despite its relative point source character the distribution of sulphur from

260 Icelandic volcanoes over the North Atlantic region was comparable to the larger scale input from

261 marine biogenic sources.

262 The most obvious and well known volcanic effect is on solar radiation, since the sulphate

263 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

288 caused a substantial modification in the aerosol chemical composition and size distribution over

289 the North Atlantic region during transport. Over the period of 11 hours the non-sea salt sulphate

290 concentration increased by a factor of four while concurrent nitrate and organic levels remained

291 low and largely unchanged. The sulphate particles were only partially neutralized. The Q-AMS

292 measurements showed that at least 35% of the total sulphate mass was pure sulphuric acid in

293 submicrometer (PM1) aerosol. Concurrent VHTDMA measurements showed that H_2SO_4

294 constituted about 26% of the total sulphate volume in particles of 50 nm diameter size. The

295 combined results suggest that there was a relatively larger degree of neutralisation by ammonium

296 in smaller particles. The predominant fraction of sulphate aerosol is assumed to have been

297 formed by aqueous phase oxidation of SO_2 based on the efficiency of this process and the cloud

298 cover associated with the air mass trajectories. A concurrent dust outbreak from Iceland

299 increased the levels of absorbing material and the light-scattering. Aerosol light scattering

300 coefficients peaked at the same time as absorption maximum and qualitatively followed sulphate

301 concentration increase. Evaluated sulphur flux demonstrated that a large amount of the total

302 (non-eruptive) sulphur emissions from Icelandic volcanoes can reach distances larger than 1000

303 km. These results in conjunction with previous research suggest that volcanogenic emissions and

304 Aeolian dust from Arctic deserts in Iceland can be potentially significant regional sources of

305 aerosols over the North Atlantic and therefore should be adequately considered in regional and

306 global climate models.

307

313

314   **References**

315   Allan J.D., Jimenez J.L., Williams P.I., Alfarra M.R., Bower K.N., Jayne J.T., Coe H., Worsnop

316   D.R., 2003. Quantitative sampling using an Aerodyne aerosol mass spectrometer 1. Techniques

317   of data interpretation and error analysis. Journal of Geophysical Research 108 (D3), 4090–4100.

318   Arnalds O., Gisladottir F. O., Sigurjonsson H., 2001. Sandy deserts of Iceland: an overview.

319   Journal of Arid Environments 47, 359–371

320   Berresheim H., Jaeschke W., 1983. The contribution of volcanoes to the global atmospheric

321   sulfur budget, Journal of Geophysical Research 88, 3732-3740.

322   Biraud S., Ciais P., Ramonet M., Simmonds P., Kazan V., Monfray P., O'Doherty S., Spain T. G.,

323   Jennings S. G., 2000. European greenhouse gas emissions estimated from continuous

324   atmospheric measurements and radon 222 at Mace Head, Ireland. Journal of Geophysical

325   Research 105(D1), 1351–1366.

326   Bott A., 1999. A numerical model of the cloud-topped planetary boundary-layer: cloud

327   processing of aerosol particles in marine stratus. Environmental Modelling and Software 14,

328   635-643.

329   Charlson R. J., Lovelock J. E., Andreae M. O., Warren S. G., 1987. Oceanic phytoplankton,

330   atmospheric sulphur, cloud albedo and climate. Nature 326, 655–661.

331   Derimian Y., Karnieli A., Kaufman Y. J., Andreae M. O., Andreae T. W., O. Dubovik, W.

332   Maenhaut, I. Koren, and B. N. Holben, 2006. Dust and pollution aerosols over the Negev desert,

333   Israel: Properties, transport, and radiative effect, Journal of Geophysical Research 111, D05205,

334   doi:10.1029/2005JD006549.

335   Draxler R.R., Hess G.D., 1997. Description of the Hysplit_4 modeling system. NOAA Tech

336   Memo ERL ARL-224, 24p. (http://www.arl.noaa.gov/ready/hysplit4.html)

337   Fitzgerald J.W., 1991. Marine aerosols: A review. Atmospheric Environment - Part A General

338   Topics 25, Issue 3-4, 533-545.

339   Fletcher C. A., Johnson G. R., Ristovski Z. D., Harvey, M., 2007. Hygroscopic and volatile

340   properties of marine aerosol observed at Cape Grim during the P2P campaign. Environmental

341   Chemistry 4, 162-171.

342   Gilliland, R. L., 1982. Solar, volcanic, and CO_2 forcing of recent climatic changes, Climatic

343   Change 4, 111–131.

344   Glasow R., Bobrowski N., Kern C., 2008. The effects of volcanic eruptions on atmospheric

345   chemistry. Chemical Geology, Article in Press.

346   Graf H.-F., Feichter J. , Langmann B., 1997. Volcanic sulfur emissions: Estimates of source

347   strength and its contribution to the global sulfate distribution, Journal of Geophysical Research

348   102(D9), 10,727–10,738.

349   Textor C., Graf H. F., Timmreck C., and Robock A., 2004. Emissions from volcanoes. In

350   Emissions of Atmospheric Trace Compounds, Claire Granier, Paulo Artaxo, and Claire Reeves,

351   Eds., (Kluwer, Dordrecht), 269-303.

352   Halmer M.M., Schmincke H.U., Graf H.F., 2002. The annual volcanic gas input into the

353   atmosphere, in particular into the stratosphere: a global data set for the past 100 years. Journal of

354   Volcanology and Geothermal Research 115, 511-528.

355   Hegg D.A., 1985. The importance of liquid phase oxidation of SO_2 in the troposphere. Journal

356   of Geophysical Research 90, 3773-3779.

357   Hoppel W. A., Frick G. M., Larson R. E., 1986. Effect of nonprecipitating clouds on the aerosol

358   size distribution in the marine boundary layer, Geophysical Research Letters 13, 125–128.

359 Ichoku C., Andreae M.O., Andreae T.W., Meixner F.X., Schebeske G., Formenti P., Maenhaut

360 W., Cafmeyer, J., Ptasinski, J., Karnieli, A., Orlovsky L., 1999. Interrelationships between

361 aerosol characteristics and light scattering during late winter in an Eastern Mediterranean arid

362 environment, Journal of Geophysical Research 104(D20), 24,371–24,393.

363 Jayne J.T., Leard D.C., Ahang X., Davidovits P., Smith K.A., Kolb C.E., Worsnop D.R., 2000.

364 Development of an aerosol mass spectrometer for size and composition analysis of submicron

365 particles. Aerosol Science and Technology 33 (1–2), 49–70.

366 Jennings S. G., Kleefeld C., O'Dowd, Junker C., Spain T. G., O'Brien P., Roddy A. F.,

367 O'Connor T. C., 2003. Mace Head atmospheric research station—Characterization of aerosol

368 radiative parameters. Boreal Environment Research 8, 303 – 314.

369 Jimenez J.L., Jayne J.T., Shi Q., Kolb C.E., Worsnop D.R., Yourshaw I., Seinfeld J.H., Flagan

370 R.C., Zhang X., Smith K.A., Morris J.W., Davidovits P., 2003. Ambient aerosol sampling using

371 the Aerodyne Aerosol Mass Spectrometer. Journal of Geophysical Research 108 (D7), 8245–

372 8258.

373 Johnson N. and Parnell R. A., 1986. Composition, distribution and neutralization of 'acid rain'

374 derived from Masaya volcano, Nicaragua. Tellus 38B, 106–117.

375 Johnson G. R., Ristovski Z., Morawska L., 2004. Method for measuring the hygroscopic

376 behaviour of lower volatiliy fractions in an internally mixed aerosol. Journal of Aerosol Science

377 35, 443-455.

378 Johnson G. R., Ristovski Z. D., D'Anna B., Morawska L., 2005. Hygroscopic behaviour of

379 partially volatilised coastal marine aerosols using the volatilisation and humidification tandem

380 differential mobility analyser technique. Journal of Geophysical Research 110, D20203.

381 Kettle A. J., Andreae M. O., 2000. Flux of dimethylsulfide from the oceans: A comparison of

382 updated data sets and flux models, Journal of Geophysical Research 105, 26,793–26,808.

383 Kim K. W., He Z., Kim Y. J., 2004. Physicochemical characteristics and radiative properties of

384 Asian dust particles observed at Kwangju, Korea, during the 2001 ACE-Asia intensive

385 observation period, Journal of Geophysical Research 109, D19S02, doi:10.1029/2003JD003693.

386 Lack D., Cappa C., Covert D., Baynard T., Massoli P., Sierau B., Bates T., Quinn P., Lovejoy E.,

387 Ravishankara A. R. Bias in Filter-Based Aerosol Light Absorption Measurements Due to

388 Organic Aerosol Loading: Evidence from Ambient Measurements (2008) Aerosol Science and

389 Technology, 42:1033–1041.

390 Mather T. A., Allen A. G., Oppenheimer C., Pyle D. M., McGonigle A. J. S., 2003. Size-

391 Resolved Characterisation of Soluble Ions in the Particles in the Tropospheric Plume of Masaya

392 Volcano, Nicaragua: Origins and Plume Processing. Journal of Atmospheric Chemistry 46, 207–

393 237.

394 Matthew B.M., Middlebrook A.M., Onasch T.B., 2008. Collection Efficiencies in an Aerodyne

395 Aerosol Mass Spectrometer as a Function of Particle Phase for Laboratory Generated Aerosols.

396 Aerosol Science and Technology 42(11), 884-898, DOI: 10.1080/02786820802356797

397 McMurry P. H., Takano H., Anderson G. R., 1983. Study of the Ammonia (Gas)- Sulphuric Acid

398 (Aerosol) Reaction Rate. Environmental Science and Technology 17, 347-352.

399 Messager C., Schmidt M., Ramonet M., Bousquet P., Simmonds P., Manning A., Kazan V.,

400 Spain G., Jennings S. G., Ciais P., 2008. Ten years of CO2, CH4, CO and N2O fluxes over

401 Western Europe inferred from atmospheric measurements at Mace Head, Ireland. Atmospheric

402 Chemistry and Physics Discussions 8, 1191–1237.

403 Middlebrook A. Bahreini R., 2008. Applying Laboratory Collection Efficiencies to Ambient

404 Field Data. AMS Users' Meeting 2008. Accessed on 18.02.2009

405 http://cires.colorado.edu/jimenez-group/UsrMtgs/UsersMtg9/09_Middlebrook_CE.pdf

406   Miller R.L., Tegen I., Perlwitz J., 2004. Surface radiative forcing by soil dust aerosols and the

407   hydrologic cycle. Journal of Geophysical Research 109 (D4), D04203.

408   Minnis, P., E. F. Harrison, L. L. Stowe, G. G. Gison, F. M. Denn, D. R. Doelling, and W. L.

409   Smith Jr., 1993. Radiative climate forcing by the Mount Pinatubo eruption, Science259, 1411–

410   1415.

411   NASA (2008). Accessed on 11/05/2009.

412   http://earthobservatory.nasa.gov/NaturalHazards/view.php?id=18595&oldid=14345.

413   O'Connor T.C., Jennings S.G., O'Dowd C.D., 2008. Highlights of fifty years of atmospheric

414   aerosol research at Mace Head, Atmospheric Research 90, 338-355.

415   O'Dowd C.D., Smith M.H., 1993./ Physicochemical properties of aerosols over the northeast

416   Atlantic: evidence for wind-speed-related submicron sea-salt aerosol production. Journal of

417   Geophysical Research 98 (D1), 1137-1149.

418   O'Dowd C. D., Low, J. A., Smith M. H., 2000. The effect of clouds on aerosol growth in the

419   rural atmosphere. Atmospheric Research 54, 4, 201-221.

420   Polian G., Lambert G., Ardouin B., and Jegou A., 1986. Long-range transport of continental

421   radon in subantarctic and antarctic areas, Tellus 38B, 178–189.

422   Prospero J. M., Arnalds Ó., Olafsson H., Bullard J. E., Hodgkins R., 2008. Iceland Dust Storms

423   Linked to Glacial Outwash Deposits and to Sub-Glacial Flood (Jökulhlaup) Events, Eos Trans.

424   AGU 89 (53), Fall Meeting Supplementary, Abstract A13E-08

425   Rampino, M. R., and S. Self, 1984. Sulphur-rich volcanic eruptions and stratospheric aerosols,

426   Nature, 310, 677–679.

427   Robock A., 2000. Volcanic eruptions and climate. Reviews of Geophysics 38, 191–219.

428   Satsumabayashi H., Kawamura M., Katsuno T., Futaki K., Murano K., Carmichael G. R., Kajino

429   M., Horiguchi M., Ueda H., 2004. Effects of Miyake volcanic effluents on airborne particles and

430   precipitation in central Japan, Journal of Geophysical Research 109, D19202.

431   Savoie D. L., Arimoto R., Keene W. C., Prospero J. M., Duce R. A., Galloway J. N., 2002.

432   Marine biogenic and anthropogenic contributions to non-sea-salt sulfate in the marine boundary

433   layer over the North Atlantic Ocean, Journal of Geophysical Research 107(D18), 4356

434   Stohl A., Forster C., Frank A., Seibert P. and Wotawa G., 2005. Technical Note: The Lagrangian

435   particle dispersion model FLEXPART version 6.2. Atmospheric Chemistry and Physics 5, 2461-

436   2474. http://niflheim.nilu.no/~burkhart/EUCAARI/MHD/MHD_200706/

437   Stothers r. B., Rampino M. R., 1983. Historic Volcanism, European Dry Fogs, and Greenland

438   Acid Precipitation, 1500 B.C. to A.D. 1500. Science, New Series, Vol. 222, 4622, 411-413.

439   Tang I. N., Munkelwitz H. R., Davis J. G., 1978. Aerosol Growth Studies- IV. Phase

440   Transformation of Mixed Salt Aerosols in a Moist Atmosphere. Journal of Aerosol Science 9,

441   505-511.

442   Tang I. N., 1996. Chemical and size effects of hygroscopic aerosols on light scattering

443   coefficients. Journal of Geophysical Research 101, 19,245–19,250.

444   Tomlinson J. M., Li R., Collins D. R., 2007. Physical and chemical properties of the aerosol

445   within the southeastern Pacific marine boundary layer. Journal of Geophysical Research 112,

446   D12211, doi:10.1029/2006JD007771.

447   Toon, O. B., and Pollack J. B., 1980. Atmospheric aerosols and climate, American Scientist 68,

448   268–278.

449   Yoon Y. J., Ceburnis D., Cavalli F., Jourdan O., Putaud J. P., Facchini M. C., Decesari S., Fuzzi

450   S., Jennings S. G., O'Dowd C. D., 2007. Seasonal characteristics of the physico-chemical

451 properties of North Atlantic marine atmospheric aerosols. Journal of Geophysical Research 112,

452 D04206; doi: 10.1029/2005JD007044.