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Impact of volcanic ash plume aerosol on cloud microphysics

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ABSTRACT

This study focuses on the dispersion of the Eyjafjallajökull volcanic ash plume over the west of Ireland, at the Mace Head Supersite, and its influence on cloud formation and microphysics during one significant event spanning May 16th and May 17th, 2010. Ground-based remote sensing of cloud microphysics was performed using a K_a-band Doppler cloud RADAR, a LIDAR-ceilometer and a multi-channel microwave-radiometer combined with the synergistic analysis scheme *SYRSOC* (*Synergistic Remote Sensing Of Cloud*). For this case study of volcanic aerosol interaction with clouds, cloud droplet number concentration (CDNC), liquid water content (LWC), and droplet effective radius (r_{eff}) and the relative dispersion were retrieved. A unique cloud type formed over Mace Head characterized by layer-averaged maximum, mean and standard deviation values of the CDNC, r_{eff} and LWC: $N_{max} = 948\ cm^{-3}$, $\overline{N} = 297\ cm^{-3}$, $\sigma_N = 250\ cm^{-3}$, $r_{eff\ max} = 35.5\ \mu m$, $\overline{r_{eff}} = 4.8\ \mu m$, $\sigma_{reff} = 4.4\ \mu m$, $LWC_{max} = 0.23\ g\ m^{-3}$, $\overline{LWC} = 0.055\ g\ m^{-3}$, $\sigma_{LWC} = 0.054\ g\ m^{-3}$, respectively. The high CDNC, for marine clean air, were associated with large accumulation mode diameter (395 nm) and a hygroscopic growth factor consistent with sulphuric acid aerosol, despite being almost exclusively internally mixed in submicron sizes. Additionally, the Condensation Nuclei (CN, $d > 10$ nm) to Cloud Condensation Nuclei (CCN) ratio, CCN:CN ~1 at the moderately low supersaturation of 0.25%. This case study illustrates the influence of volcanic aerosols on cloud formation and microphysics and shows that volcanic aerosol can be an efficient CCN.

1. Introduction

On March the 20th, 2010, the Icelandic Eyjafjallajökull volcano (63°38′0″ N, 19°36′0″ W, summit elevation 1666 m) erupted after almost 190 years since its last most significant eruption occurred between 1821 and 1823. Although the eruption of 20 March has been classified only as 1 on the Volcanic Explosivity Index (Newhall and Self, 1982; Zehner, 2010), in the following weeks the seismic activity, already started at the end of 2009, became more intense with large amount of eruptive material emitted into the lower Troposphere. The eruption of 14 April 2010 created an ash cloud consisting mainly of tephra produced by thermal contraction from contact with the melted ice cap; the explosively ejected material that formed the ash cloud had diverse size distribution ranging from the fine <100 μm to the coarse ash <2 mm. Larger particles like lapilli and pyroclastic material fell out of the cloud in the first minutes to hours after being ejected from the vent. In the first days after the eruption the fine-ash cloud has been transported by Westerlies out to northern Europe first and to central and southern Europe thereafter. The across-Europe and North Atlantic-transported ash cloud led, under the directions of the London Volcanic Ash Advisory Centre, to the closure of most of the European airspaces. The scientific community was committed to assess the actual potentialities of in-situ and ground/space-based remote sensor measurements to detect the ash plume and to retrieve its density and size distribution. Most of the remote sensing measurements focused on the physical and optical properties of the ash plume leaving little focus to the actual interaction of the ash particles with clouds. This study presents a case of cloud formation directly from the volcanic ash layer advected over the GAW atmospheric station of Mace Head, Ireland, on the night between 16 and 17 May 2010.

The mechanisms regulating the formation of clouds have been studied for almost one hundred years now, but it is only with the onset of the recent technologies that it has been possible to investigate the microphysics of the aerosols–cloud interaction. Although numerous studies have improved the knowledge of the aerosol indirect effects (Twomey, 1977; Ackerman et al., 2000; Rosenfeld et al., 2006), much more needs to be learned on the mechanisms leading to cloud formation from a given cloud condensation nuclei (CCN) distribution. The naturally produced CCN in the atmosphere depend strongly on the spatial distribution of several gases and particles, namely phytoplankton emitted dimethyl-sulphide (DMS), oxidised SO_2, MSA, H_2SO_4, SO_4, organics and dust particles. The sensitivity of the activated cloud droplets to the different populations

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of CCN is a complex process (Woodhouse et al., 2010) which depends mainly on aerosol size, hygroscopicity and on the local values of temperature and supersaturation with respect to water or ice or both depending on the cloud phase. Clouds stemming from dust or ash CCN are not uncommon although they are rarely observed due to narrow conditions of temperature and humidity in which they form in combination with such CCN (Flamant et al., 2009; Bou Karam et al., 2009). In polluted areas, a large number of aerosols serving as CCN would lead to increased cloud droplet number concentrations (CDNC). However, embryonic and fully-developed CDNC would form in conditions of low per-droplet available water than in clean conditions. The more numerous cloud droplets will have smaller size by 20–30% increasing the albedo by almost the 25% (Kaufman et al., 2002). Smaller cloud droplets are less efficient in generating precipitation and this may affect the local hydrologic cycle. Nevertheless, the way a population of many large aerosol particles can influence the mechanism of cloud formation and ultimately affect the precipitation process is not well understood yet. The radiative aerosols first and second indirect effects are still suffering from a larger uncertainty compared to the already large uncertainty related to the direct effect by the greenhouse gases. This study is intended to provide a quantitative microphysical analysis of the aerosol indirect effect for a case of volcanic ash CCN.

2. Site and instruments

2.1. The site

Located on the west coast of Ireland, the Atmospheric Research Station at Mace Head, Carna, County Galway is unique in Europe in that its location offers westerly exposure to the North Atlantic Ocean through the clean sector (190°–300°N) and the opportunity to study atmospheric composition under Northern Hemispheric background conditions as well as European continental emissions when the winds favour transport from that region (O'Connor et al., 2008). The site location, at 53° 20 min N, 9° 54 min W, is in the path of the mid-latitude cyclones which frequently traverse the North Atlantic. The instruments are located 300 m from the shore line on a gently-sloping hill (4° incline).

2.2. Instruments

2.2.1. Remote sensors

The GAW Atmospheric Station of Mace Head is part of the CLOUDNET programme (Illingworth et al., 2007) since 2008 using synergistic input data from three remote sensors to supply the CLOUDNET modules. In parallel to CLOUDNET the authors have developed the SYRSOC (*SY*nergistic *R*emote *S*ensing *O*f *C*loud) technique (see Appendix A for a complete description and work by Martucci and O'Dowd, 2011) to retrieve the full cloud microphysics from the three remote sensors, namely the Jenoptik CHM15K LIDAR ceilometer with 1064-nm wavelength and 15-km vertical range (Flentje et al., 2010; Martucci et al., 2010a), the RPG-HATPRO water vapour and oxygen multi-channel microwave profiler (Lönhert et al., 2009) and the MIRA36, 35 GHz K_a-band Doppler cloud RADAR (Melchionna et al., 2008).

2.2.2. Chemical, physical and optical instruments

The size resolved non-refractory chemical composition of submicron aerosol particles was measured with an Aerodyne High Resolution Time of Flight Aerosol Mass Spectrometer (HR-ToF AMS, Aerodyne, Billerica, MA). Detailed instrument description could be found in the work from DeCarlo et al. (2006). HR-ToF AMS was routinely calibrated according to the methods 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. Considering particle chemical composition and relative humidity (Matthew et al., 2008; Middlebrook and Bahreini, 2008) a collection efficiency of $C = 1$ was applied for the measurements discussed in this study.

The size distribution of ambient particles sampled few meters above the ground level was performed in parallel by two instruments, the scanning mobility particle sizers (SMPS) over a particle diameter range from 3 nm to about 0.5 μm, and the Aerodynamic Particle Sizer (APS) model 3021 (TSI Inc) with 51 channel of equal logarithmic width of 0.03 within the size range of 0.3–20.0 μm.

Aerosol scattering measurements by means of a three-wavelength integrating Nephelometer (TSI Model 3551) are conducted routinely at Mace Head. Aerosol absorption was measured using a Multi-Angle Absorption Photometer (MAAP).

3. Case description

The ash plume emitted by the Eyjafjallajökull volcano on14 May 2010 reached the Troposphere above Mace Head ($\bar{z} = 3.7$ km a.g.l.) on the night between the 16th and 17th of May. The plume was transported from the main volcano's crater to Mace Head in about 2 days in a synoptic northwest-southeast flow. In Fig. 1a the 60-h forward-trajectory calculated by NOAA HYSPLIT model with starting time at 0800 UTC and 4000-m level, shows the Icelandic origin of the advected air mass reaching Mace Head on 16 May. Compared to the first volcanic plume observed at Mace Head on 19–20 April 2010 (i.e. 5 days after the first Eyjafjalla explosion on 14 April 2010) some major difference have been observed (O'Dowd et al, "The Eyjafjallajökull Ash Plume – Part I: Physical, Chemical and Optical Characteristics" This Issue) with respect to the discussed case of 16–17 May. Mainly two differences characterized the two events: (i) less sulphate ($\sim$2 μg m^{-3}) compared to the May-event ($\sim$7 μg m^{-3}) most likely due to cloudless conditions on the 19–20 April and (ii) more diverse size distribution of ash and overall larger sizes in April than in May (Scanning Electron Microscope analysis). Fig. 1b shows the temporal evolution of the spatial distribution of the volcanic plume simulated by the REMOTE (REgional MOdel with Tracer Extension) regional climate model (Langmann, 2000; Varghese et al., 2011; O'Dowd et al., 2012b) for 16 May at 1500 GMT to 17 May at 0600 GMT using ECMWF analysis boundary data. The REMOTE model is coupled with state-of-the-art atmospheric chemistry and aerosol dynamics modules and is capable of precise estimation of volcanic ash distribution.

Fig. 2 shows the time-height cross section of the logarithmic range-corrected LIDAR signal normalized by the LIDAR ratio and the instrumental LIDAR constant. The timeseries starts at 1900 UTC on 16 May and shows the volcanic plume in four highlighted areas between 3 and 6 km a.g.l.; the volcanic plume is separated into three layers, between 5.5 and 6 km, between 3.7 and 4.6 km and at $\sim$3 km a.g.l. Only the middle layer which subsides from $\sim$4.6 (1930 UTC) to 3.7 km a.g.l. (2230 UTC) contributes to the cloud formation. Boundary-layer stratus clouds at $\sim$1.5 km a.g.l. prevent the LIDAR to detect further above the cloud layer during 1945–2210 UTC. By the time the boundary-layer stratus clouds dissolved, the plume at 3.7 km a.g.l. became visible for other 20 min (the lower plume layer at $\sim$3 km a.g.l. remained visible till midnight). At 2230 UTC a cloud formed from the plume at 3.7 km a.g.l. and lasted 3.5 h (till 26 UTC on the *x*-axis of Fig. 2) before evaporating and leaving the volcanic plume still at 4 km a.g.l. A new cloud event formed from the volcanic plume at around 0350 UTC (2650 UTC on the *x*-axis of Fig. 2) at almost the same height.

The cloud observed from 2230 UTC to 0210 UTC on the following day stemmed from two populations of sulphate-coated CCN with accumulation mode at 390 nm and Aitken mode at 90 nm which