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# The Eyjafjallajökull eruption in April 2010 – detection of volcanic plume using in-situ measurements, ozone sondes and a new generation ceilometer network

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## Abstract

Volcanic emissions from the Eyjafjallajökull volcano on the Southern fringe of Iceland were detected at different places in Germany by means of in-situ measurements, ozone sondes and a dense network of ceilometers of the German Meteorological Service (DWD). In-situ measurements of sulphur dioxide and particulate matter were performed at the Global Atmosphere Watch (GAW) station Zugspitze/Hohenpeissenberg. At Hohenpeissenberg, a number of reactive gases, e.g. sulphuric acid, carbon monoxide and nitrogen oxides were additionally measured during the period of interest. Also ozone sondes were launched at Hohenpeissenberg in the pre-alpine area. A newly established network of ceilometers (Jenoptik CHM15K) at currently 36 meteorological stations in Germany provided the temporal evolution of emissions over Germany. The sensitivity of these instruments with respect to atmospheric aerosols further allowed the inversion of gathered backscatter profiles, and aerosol extinction coefficients and particle mass concentration were finally obtained.

## 1 Introduction

After months of enhanced seismic activity and crustal deformation close to the volcano summit, the Eyjafjallajökull volcano located in the south of Iceland (63°38'0" N, 19°36'0" W, summit 1660 m) erupted on 20 March 2010; it was the first major outbreak after a relatively silent period of nearly 190 years. Fire fountains, lava jets and eruptions were visually observed but volcanic emissions remained in the lower atmosphere. A major outbreak of the central crater under the covering ice cap followed on 14 April 2010 (IES, 2010). The eruption was enforced when water from the melting ice cap of the summit and surrounding glaciers met the lava. A large volcanic ash plume rising up to the tropopause level (~10 km) was observed by satellite instruments.

A high pressure system over Iceland on 16 April and later Western Scandinavia favored with north-westerly winds the transport of large amounts of erupted material,

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typically volcanic ash, water vapour, and sulphur dioxide (SO<sub>2</sub>), across the North Atlantic towards the British Islands, Scandinavia and later on to Central Europe. Such plumes may heavily affect aircrafts (Miller and Casadevall, 2000) and the detection and tracking of plumes by satellites for aviation hazard mitigation supports the work of Volcanic Ash Advisory Centers (VAAC) (Carn et al., 2008; Prata, 2008). The London VAAC being responsible for the Northern European sector including Iceland issued warnings about the ash plume and consequently the majority of European airports were closed from 15 April to 21 April 2010. Since then the Eyjafjallajökull showed variable activity and series of eruptions caused further volcanic emissions that reached the British Islands, the Iberian peninsula, the Azores, and Italy interrupting air traffic again, e.g. from 5 May to 8 May 2010, 10 May to 12 May 2010.

The first eruptive period of the Eyjafjallajökull between 14 April and 19 April caused volcanic ash plumes reaching Northern Germany between 2 and 7 km altitude during the night from 15 April to 16 April; it subsided and spread southward on 16 and 17 April. By noon of 17 April it was entrained into the planetary boundary layer. The air mass was further deflected towards west and east at the Alps and resided over Central Europe for several days thereafter.

We describe in this study the temporal evolution of this event, using collocated in-situ measurements of trace gases and aerosol particles, ozone sondes and a newly established network of ceilometers, typically used for cloud base height detection, that was used here for aerosol profiling.

## 2 Measurements of volcanic emissions

### 2.1 The DWD ceilometer network

The spatial and temporal evolution of volcanic emissions over Germany was observed by the German ceilometer network operated by the German Weather Service (DWD) described in detail by Flentje et al. (2010). Though originally set-up for cloud base

height monitoring, the new generation of instruments (CHM15K, Jenoptik) allows the detection of aerosol layers with extinction coefficients down to about  $5 \times 10^{-6} \text{ m}^{-1}$ . The current (spring 2010) geographical position of now 36 ceilometers in Germany is indicated in Fig. 1 by so-called curtain plots.

The CHM15K uses a diode-pumped Nd-YAG solid state laser at 1064 nm yielding about  $8 \mu\text{J/pulse}$  at a 5–7 kHz repetition rate. The pulsed laser beam (divergence  $100 \mu\text{rad}$  or about 20 s of arc) is emitted off-axis of a Newton-type receiving telescope with an aperture of again  $100 \mu\text{rad}$ . Back-scattered radiation is collimated, then filtered with a narrow-band interference filter before it is detected with an avalanche photodiode run in photon counting mode. Limited by the overlap between the laser beam and the telescope field of view, the signal can be used from about 600 m above ground level up to 15 km with a vertical resolution of 15 m.

The CHM15K ceilometers provide profiles of particle and molecular back-scattering in an atmospheric column by solving the lidar equation for the backscatter coefficient profile (e.g., Klett, 1981). It requires independent information on the backscatter-to-extinction ratio (lidar ratio, LR profile) and on the backscatter coefficient at a reference height. As mostly no height resolved particle information is available, the LR is usually taken from corresponding closure studies (e.g., Mattis et al., 2004) and regarded as height independent. The wavelength of 1064 nm provides relatively large contrast to molecular scattering, thus highlighting aerosol structures, but the scattering efficiency drops sharply for particles with radii well below  $1 \mu\text{m}$ , which limits the accuracy of a single profile.

## 2.2 In-situ measurements

In-situ measurements of volcanic emissions are typically confined to particle measurements and sulphur dioxide concentrations in ambient air. Furthermore, the chemical composition of aerosols and rainwater samples can be analyzed retrospectively (Zerefos et al., 2006). Such measurements are continuously operated by DWD at the Environmental Research Station (UFS) “Schneefernerhaus” (<http://www.schneefernerhaus.>

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de/e-ufs.htm, 47.4° N, 11.0° E 2650 m a.s.l., 300 m below the Zugspitze summit) in the Northern alpine area, and at the Hohenpeissenberg Meteorological Observatory HPB (<http://www.dwd.de/gaw>, 47.8° N, 11.0° E 985 m a.s.l., 300 m above the surrounding countryside) located in the Northern pre-alpine area of Germany (Mannschreck et al., 2004).

Sulphur dioxide is measured with UV pulsed fluorescence analyzers by Thermo Instruments Inc., (model TE43i TLE at UFS and TE43 CTL at HPB), both with detection limits of about 50 ppt (parts per trillion=picomole/mole). Furthermore, measurements (only at HPB) of sulfuric acid (H<sub>2</sub>SO<sub>4</sub>) are performed by a Chemical Ion Mass Spectrometer (CIMS) instrument involving continuous sampling of ambient air, followed by chemical ionisation with NO<sub>3</sub><sup>-</sup> ions, cluster dissociation, and mass selective detection (Berresheim et al., 2003). Sulphur dioxide in-situ measurements are performed since 1995 at the Hohenpeissenberg Observatory and since 2000 also at the “Schneefernerhaus”. CIMS observations of H<sub>2</sub>SO<sub>4</sub> started in the late nineties and are operated routinely for more than 10 years now.

Particle number concentration is measured at the UFS since 2000 by a Buthanol-based condensation particle counter (model TSI CPC 3025a). At HPB, particle measurements (total particle mass, particle number concentration, scattering coefficient) for particles larger than 3 nm are continuously available since 1995 (TSI CPC 3025a, 3762, 3772, 3776). The scattering coefficient is measured by a three wavelengths nephelometer (center wavelengths 450 nm, 550 nm, 700 nm, model TSI 3563). An optical particle counter (GRIMM-OPC) complements measurements at HPB of the particle number concentration of larger particles with diameters between 0.3 μm and 30 μm. Smaller particles (between 10 nm and 900 nm) are observed since June 2007 by a Scanning Mobility Particle Sizer (SMPS) which was recently (2009) equipped with an additional sampling channel with a thermodenuder.

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## 2.3 Ozone sondes and sulphur dioxide column measurements

The Hohenpeissenberg observatory acts as the Regional Ozone Center of the Regional Association VI (RA) of the World Meteorological Organization (WMO). As part of this activity a Brewer spectrometer is operated since 1983. These types of instruments are partly sensitive to sulphur dioxide and can be used to estimate the total atmospheric SO<sub>2</sub> content. The Brewer instruments are however designed to measure primarily the atmospheric ozone content while the retrieval of sulphur dioxide is a side effect of these measurements being necessary for correct ozone measurements. A larger uncertainty of sulphur dioxide amounts retrieved from Brewer data is therefore expected, as it is discussed in detail by Fioletov et al. (1998).

The ozone center at the Hohenpeissenberg observatory launches ozone sondes (Brewer Mast) regularly since 1967, e.g. supporting international programmes monitoring the evolution of the atmospheric ozone content. Brewer Mast ozone sondes are being coupled with Vaisala RS92 radiosondes and are normally launched 2–3 times per week. The sonde measures ozone partial pressure, temperature, pressure, humidity, windspeed and -direction every second. Geographical coordinates and geometrical altitude are provided by a GPS module. During flight the data stream is transmitted in real-time to the ground station.

## 3 Results and discussion

### 3.1 Ceilometer observations

The eruption of the Icelandic Eyjafjallajökull volcano on 14 April 2010 and later on massively injected particles into the troposphere up to about 10 km altitude. The DWD ceilometer network well tracked the Eyjafjallajökull ash layers over Germany. The Northern German ceilometer stations observed the plume first in about 7 km altitude on 16 April (Fig. 2). It further crossed Germany from North to South on the frontside of an

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extended Azores High. At the middle and southern ceilometer stations the front of the ash layer arrived in about 6–7 km height but after several hours the layer concentrated between 2–4 km altitude only, forming a dense, roughly one km thick layer. In the small figures inside Fig. 1 this appears as a down tilting red-coloured layer. Note that the seemingly intermittence of the layer at many stations is due to low clouds which prevented the observation of the ash-layer above.

On 17 April the ash layer was only observed over the mainly cloud-free parts of Southern Germany where it was entrained into the convective PBL from noon onwards (Fig. 2). The main event from 17 April till 20 April as seen by the HPB ceilometer is depicted in Fig. 3. At Hohenpeissenberg the peak extinction coefficients and the mass concentrations on 17 April were roughly estimated to be  $4(\pm 2) 10^{-4} \text{ m}^{-1}$  and  $500\text{--}750(\pm 300) \mu\text{g}/\text{m}^3$ , respectively, based on co-located aerosol optical depth and nephelometer (scattering coefficient) measurements (see details in Flentje et al., 2010).

### 3.2 Ground-based in-situ observations

The global atmospheric sulphur dioxide budget is controlled by natural emissions from volcanoes, the oceans (formation of  $\text{SO}_2$  from dimethylsulfide,  $\text{H}_2\text{S}$ ) and anthropogenic emissions (Graf et al., 1997), and the removal of  $\text{SO}_2$  by wet deposition and reaction with the OH radical mainly in the troposphere. Due to its tropospheric life time of typically several days it is an excellent tracer of long-range transport of volcanic emissions but also for anthropogenic emissions. In volcanic ash plumes, sulphur dioxide is substantially enhanced and subsequently sulphuric acid may be formed during day (photochemistry), thus giving rise to secondary particle formation. Accordingly, number concentrations in ash plums are generally enhanced due to primary ash particles ( $\mu\text{m}$  size range) and occasionally newly generated smaller particles.

Simultaneously with the arrival of the ash plume, the surface concentrations of  $\text{SO}_2$  and large particles rose from background concentrations to high levels both at the “Schneefernerhaus” and later at HPB during the morning of 17 April. The strong increase was more pronounced at the UFS where the largest  $\text{SO}_2$  mixing ratio of

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about 3.5 ppb (parts per billion) in April since the beginning of the measurements was recorded (Fig. 4). The higher SO<sub>2</sub> values at the UFS support the finding (from the ceilometer measurements) that the volcanic plume subsided from higher atmospheric levels into the planetary boundary layer (PBL). After reaching their first maximum during the night 17/18 April, SO<sub>2</sub> decreased on 18 April but exhibited a second maximum in the afternoon of 19 April. The concentrations fell back to background levels during 21 April. The particle number concentration of particles larger than 3 nm are highly correlated with SO<sub>2</sub> measurements (as well as corresponding PM<sub>10</sub> values measured by the Federal Environmental Agency UBA, personnel communication L. Ries, not shown here). Aerosol parameters and SO<sub>2</sub> show both a coincident maximum and similar temporal variation.

At HPB, 40 km north of the UFS and situated in the PBL, also clear signs of volcanic impact were observed, though partially masked by enhanced anthropogenic background. Sulphur dioxide mixing ratios rose from the afternoon of 17 April (14:30 CEST) to several consecutive peaks around 1.5 ppb on 19–22 April in five distinct maxima (see Fig. 5). Highest values of about 2.3 ppb were seen during morning hours on 21 April (Fig. 5), being higher than the 99 percentile for hourly measurements in April between 2000 and 2007. The daily pattern of anthropogenic tracers such as carbon monoxide (CO), nitrogen oxides (NO, NO<sub>y</sub>) and ozone (O<sub>3</sub>) did not show any unusual temporal changes, indicating that the enhanced SO<sub>2</sub> at HPB is not due to anthropogenic emissions. Dust events on the other side (e.g. from the Saharan) would not be associated with enhanced SO<sub>2</sub> concentrations. We therefore interpret our measurements in the sense that we indeed monitored the volcanic ash plume.

Parallel observations of the CIMS instrument at HPB allowed measurements of the ambient concentration of sulfuric acid. Enhanced levels of SO<sub>2</sub> are expected to further translate into higher values of H<sub>2</sub>SO<sub>4</sub>. Oxidation of sulphur dioxide by the OH radical is the main atmospheric sink of SO<sub>2</sub> and this process is more efficient for higher solar radiation. Associated with high SO<sub>2</sub> and intensive photochemistry, peak sulphuric acid concentrations on 19 April were above the hereto observed maxima in April (Fig. 6).

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The arrival of the plume is also observed in the number concentration of particles of 1–4  $\mu\text{m}$  diameter which represents the typical size range of aged volcanic ash plumes. The size-resolved particle number concentration measured by an optical particle counter (see above) at HPB from 13 April to 23 April is shown in Fig. 7. On 17 April the concentration of larger particles ( $>1 \mu\text{m}$ ) suddenly rises and values were enhanced for the next six days. The increasing particle number concentration of large particles is temporally in phase with the corresponding ambient  $\text{SO}_2$  enhancement. The amount of smaller particles around 0.5  $\mu\text{m}$  was not affected, as expected, since volcanic ash typically consists of larger particles. Precipitation on 18 April and 20 April led to decreasing particle concentrations. Due to enhanced sulphuric acid concentrations in volcanic ash plumes, coating of the primarily hydrophobic ash particles is expected to achieve hydrophilic surfaces. Hence, these could act as condensation nuclei or become attached to cloud drops and subsequently be washed out of the atmosphere. Thus, removal of ash particles is expected to proceed more rapidly than due to simple sedimentation alone.

Other measurements within the GAW measurement programme at HPB, the chemical analysis of rain water probes and water-soluble aerosols, support the presence of volcanic emissions as well. Enhanced concentrations and deposition of heavy metals ions (iron, manganese) were found in weekly rain water probes, although not being extraordinary high if compared to other samples during the last 1.5 years. Daily rain water probes analyzed by ion chromatography showed higher (but not unusually high) amounts of sulphate, fluorine and calcium ions indicating altogether the presence of volcanic emissions.

### 3.3 Ozone sondes and total column measurements

Ozone sondes were launched daily between 14 April and 25 April 2010 (excluding 15 April). The launch on 19 April supported a flight campaign carried out by the Falcon research aircraft of the German Aerospace Center (DLR) in the afternoon around 14:15 UTC. Little is known about the impact of volcanic ash and/or volcanic sulphur

dioxide (and precursor components) on ozone sonde measurements (Schenkel and Broder, 1982).

The retrieved ozone profile from 17 April showed a severely disturbed ozone profile in the lower troposphere between 2.5 km and 7 km a.s.l. (Fig. 8). Layers with reduced ozone appeared also during the following days, but not as pronounced as on Saturday, 17 April. In fact, such a distorted profile was never recorded before in the more than 40 years history of ozone sondes launched at Hohenpeissenberg. Also, the ozone sonde launched by the Koninklijk Nederlands Meteorologisch Instituut (KNMI) at DeBilt on 16 April (one day earlier) showed a similar pattern in the lower troposphere between 2 km and 5 km (V. Huijnen, pers. communication). At that time the ash plume arrived from north-westerly direction at the Netherlands and Northern Germany. Part of the registered low ozone values maybe caused by SO<sub>2</sub> interfering with the wet-chemical measurement: 1 ppb SO<sub>2</sub> is registered as -1 ppb O<sub>3</sub> (Schenkel and Broder, 1982). Effects of volcanic ash on the measurement can also not be excluded. It is however plausible that substantial ozone amounts were destroyed in the Eyjafjallajökull plume. Enhanced ozone depletion was also observed by aircraft measurements in the plume on 3 May 2010 (Schumann et al., 2010). Figure 8 shows the last sounding before arrival of the plume, the sounding through the main plume, and two later soundings. For comparison, the white and grey areas show the range of ozone values observed in April 2007 to 2009.

The SO<sub>2</sub> observations at the UFS coincide further with the SO<sub>2</sub> columnar measurements of the Brewer spectrometer at Hohenpeissenberg (Fig. 9). The Brewer spectrometer detected a columnar content of 0.7 DU (Dobson units) on 17 April (daily mean). This corresponds to a layer of about 3 km thickness at a concentration of 3 ppb SO<sub>2</sub>, or a 1 km layer at 10 ppb SO<sub>2</sub>. The latter concentration/layer combination better corresponds to the thickness of the ash layer as derived from the Hohenpeissenberg ceilometer, while the former combination better supports the concentration measured at the UFS. Differences between the thickness of the ash layer and the corresponding SO<sub>2</sub> layer could be due to different sedimentation processes. The ash plume arrived

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later at the UFS and it therefore appears plausible that the volcanic emissions have started to interfere and mix into the planetary boundary layer causing reduced concentrations. The “Schneefernerhaus” might have been additionally influenced by upslope winds with potentially lower SO<sub>2</sub> mixing ratios. A second relative maximum of the SO<sub>2</sub> column of about 0.4 DU was observed on 20 April which corresponds to enhanced SO<sub>2</sub> levels in ambient air at Hohenpeissenberg. The low total SO<sub>2</sub> columns on 18 April and 19 April are probably due to precipitation at Hohenpeissenberg during the evening hours on 18 April (1.8 mm).

## 4 Conclusions

Our study shows that measurements collected at the GAW global station Zugspitze/Hohenpeissenberg and data from Germany’s national ceilometer network can be used together to describe the spatial and temporal evolution of the Eyjafjallajökull ash plume that passed Central Europe from 16 April to 23 April 2010. We conclude that combined observations of SO<sub>2</sub> and particles backed by measurements of anthropogenic trace gases (CO, NO, NO<sub>2</sub>) form a unique tracer to detect volcanic ash layers in the atmosphere. Other sources of enhanced particle concentrations due to anthropogenic emissions and dust could be ruled out by missing coincident increases of CO and nitrogen oxides in case of anthropogenic plumes.

The ceilometer network detected the ash plume first over Northern Germany. Within one day and being qualitatively in line with model predictions the plume crossed Germany from North-West to South-East and various in-situ measurements responded to the changed atmospheric composition. Although error bars seem large, the uncertainties are small enough to make use of the network for aviation advisories, indicating whether the legal flight ban threshold of presently 4 mg/m<sup>3</sup> is imminent to be exceeded.

The particle mass concentrations retrieved are comparable to in-situ measurements gathered during a dedicated measurement flight (B. Weinzierl, German Aerospace Center DLR, personal communication). The ceilometer data fit also well into all the

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other in-situ measurements gathered at HPB and UFS. Simultaneous increases of the  $\text{SO}_2$  and  $\text{H}_2\text{SO}_4$  concentrations as well as the number concentration of larger particles ( $d > 1\text{--}4\ \mu\text{m}$ ) and the chemical composition of rain water confirm the volcanic origin of the analysed layers.

5 Under clear sky conditions, optical remote sensing instruments from the ground can retrieve the vertical distributions of volcanic ash (ceilometers, aerosol lidars, MAX-DOAS) or the total column in case of  $\text{SO}_2$  (Brewer, MAXDOAS). For an unquestionable identification of volcanic ash plumes, however, in-situ measurements of such layers from high mountain stations and aircrafts, where concentrations of the volcanic ash  
10 can be determined directly, are highly recommended. High-altitude observations also allow measurements when low clouds hinder the use of optical remote sensing instruments from ground stations.

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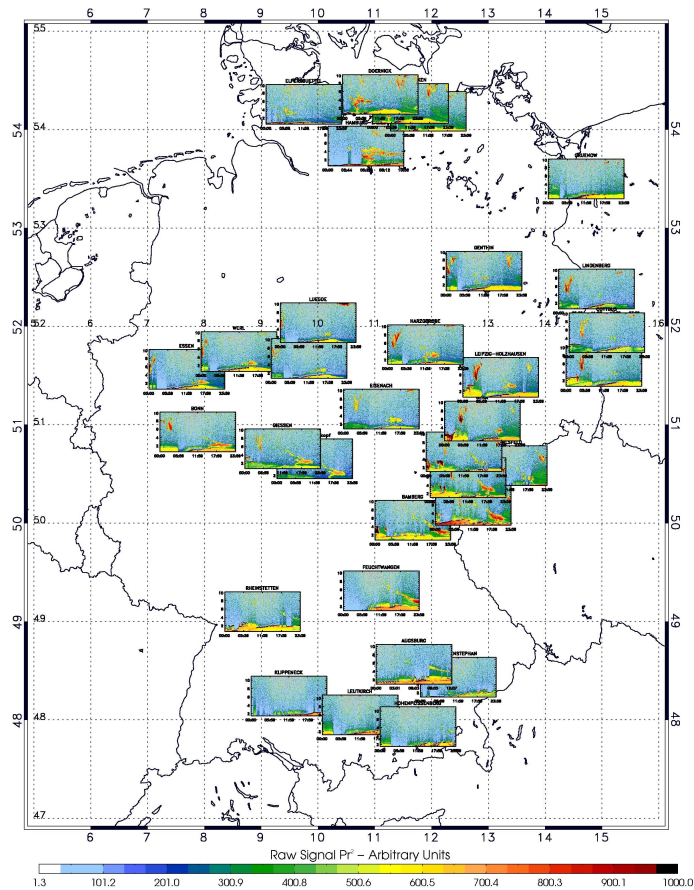
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**Fig. 1.** Geographical distribution of the DWD ceilometer network as of spring 2010. Curtain plots of diurnal backscatter vertical profiles (from 16 April) indicate the position of the measurement devices.

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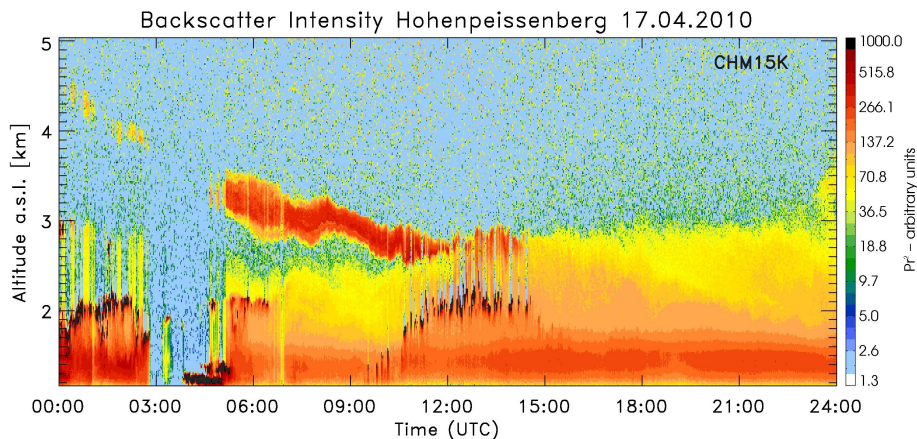
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**Fig. 2.** Backscatter profiles obtained from the Hohenpeissenberg ceilometer on 17 April. Clearly the arrival of the ash plume during morning hours in about 3–4 km is visible, although low-level clouds weakened the corresponding backscatter signal. The entrainment of the ash plume into the planetary boundary layer starts around local noon.

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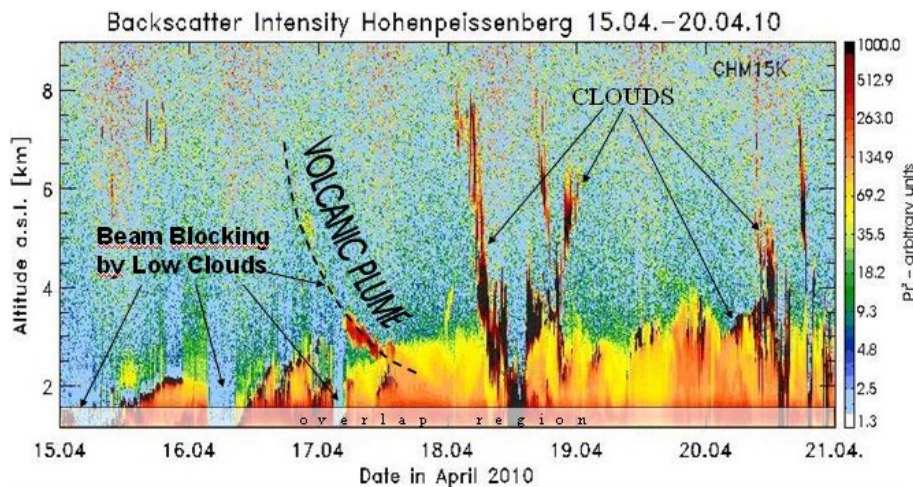
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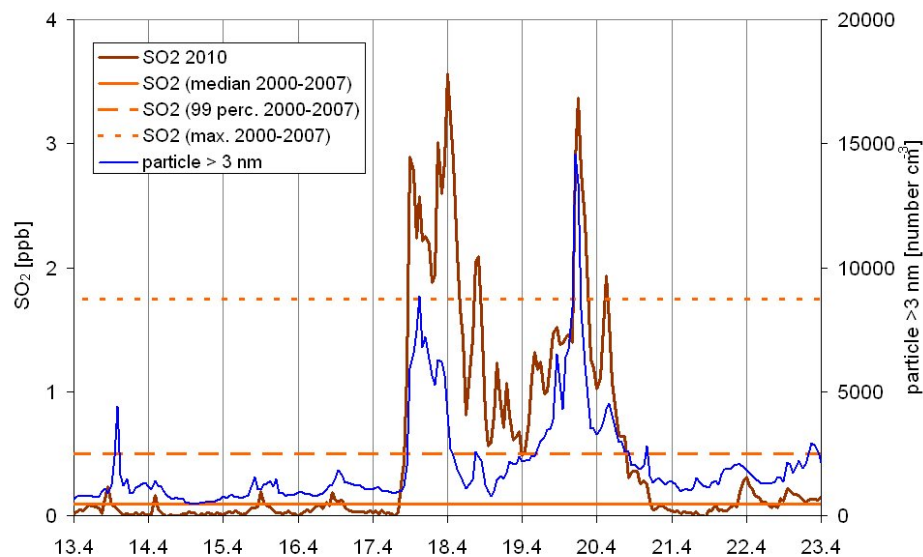


**Fig. 3.** Time series of backscatter profiles of the Hohenpeissenberg ceilometer from 15 April to 20 April 2010. Clouds are often hampering the observation of higher tropospheric layers while the arrival of the ash plume could be observed during a mostly cloud-free period.



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**Fig. 4.**  $\text{SO}_2$  mixing ratios and particle number concentrations ( $>3$  nm) measured by DWD at the GAW Global Station Zugspitze/Hohenpeissenberg, platform Zugspitze/UFS at 2650 m elevation. The particle number concentrations are highly correlated with  $\text{SO}_2$ .

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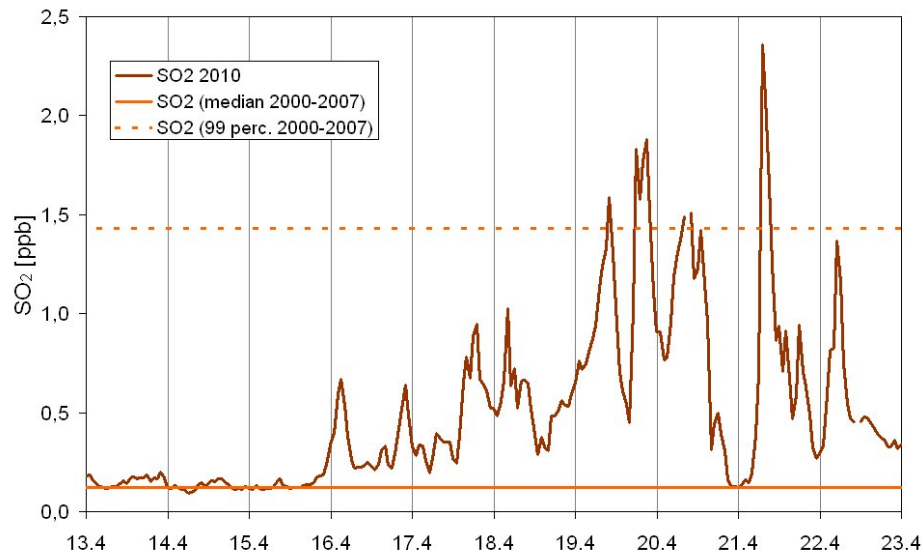
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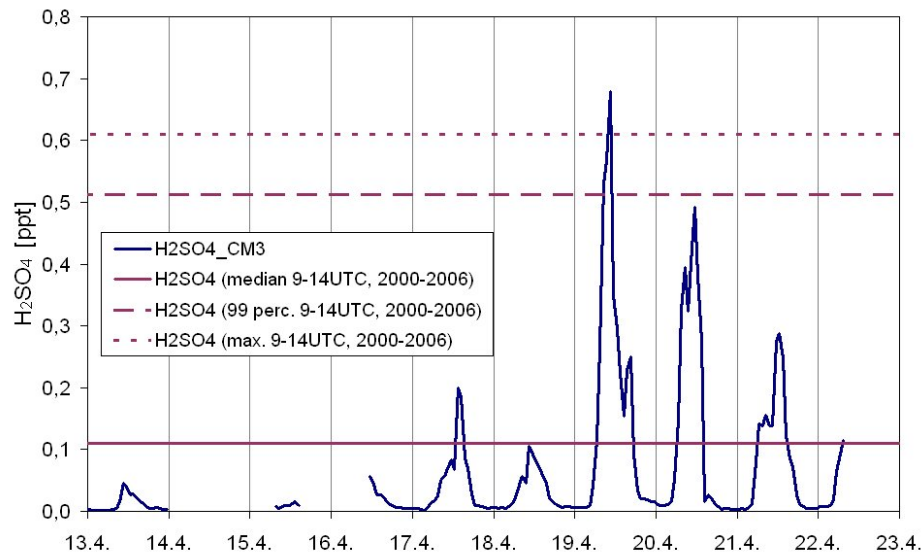
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**Fig. 5.** SO<sub>2</sub> mixing ratio measured at the GAW Global Station Zugspitze/Hohenpeissenberg, platform HPB at 980 m elevation.

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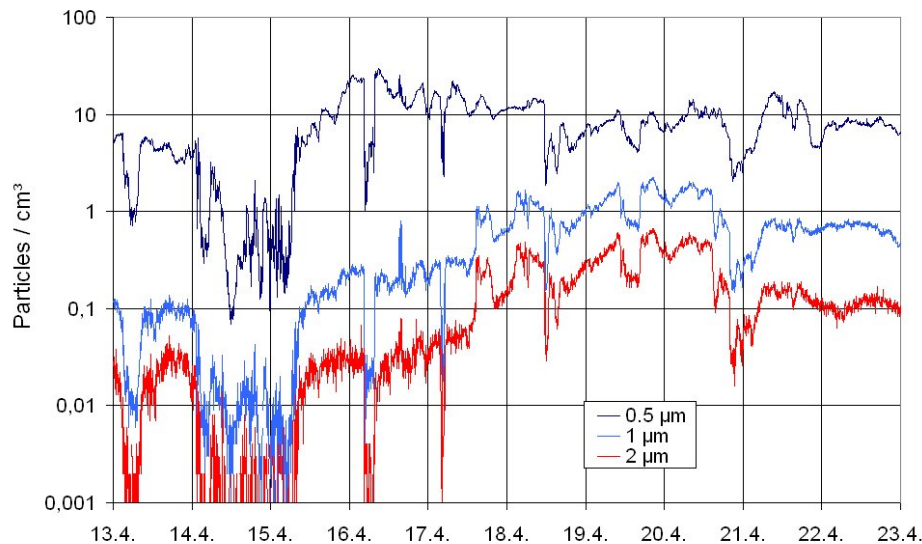


**Fig. 6.** Sulfuric acid ( $\text{H}_2\text{SO}_4$ ) measurements at the GAW Global Station Zugspitze/Hohenpeissenberg, platform HPB at 980 m elevation as derived from a Chemical Ion Mass Spectrometer (CIMS).

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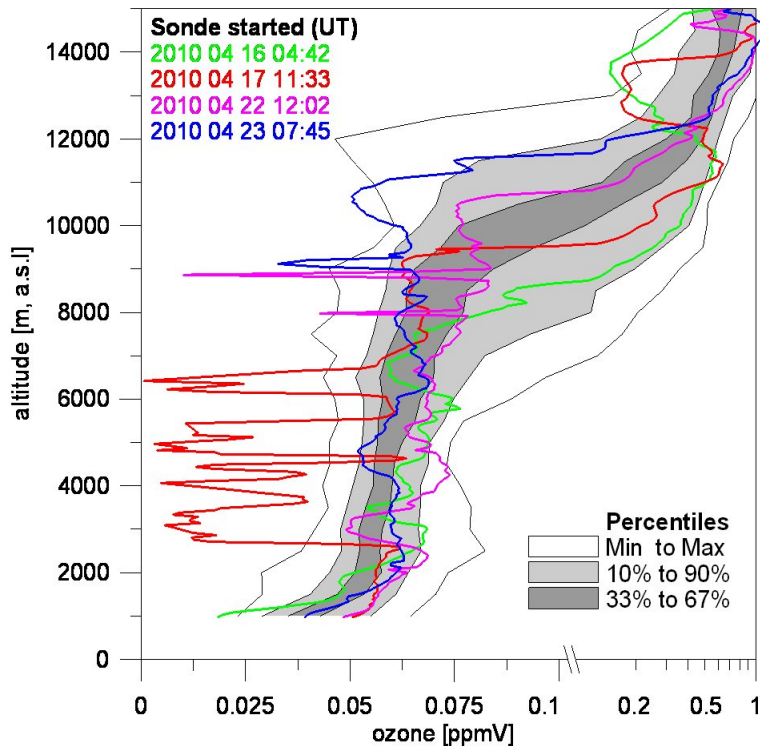
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**Fig. 7.** Size-resolved particle number concentration at the GAW Global Station Zugspitze/Hohenpeissenberg, platform Hohenpeissenberg at 980 m elevation as derived from an optical particle counter (GRIMM-OPC).

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**Fig. 8.** Ozone profiles derived from ozone sondes launched at Hohenpeissenberg between 14 April and 25 April. On 17 April the ozone content between 2.5 km and 7 km is largely reduced within several distinct layers (red curve). Grey-shaded areas show the 10–90 and 33–67 percentiles of all April measurements of years 2007 to 2009, respectively. An undisturbed ozone profile was observed on 14 April (green) while the ozone profiles of 22 April (magenta) and 23 April (blue) showed layers with sharply dropped ozone concentrations between 8 km and 9 km.

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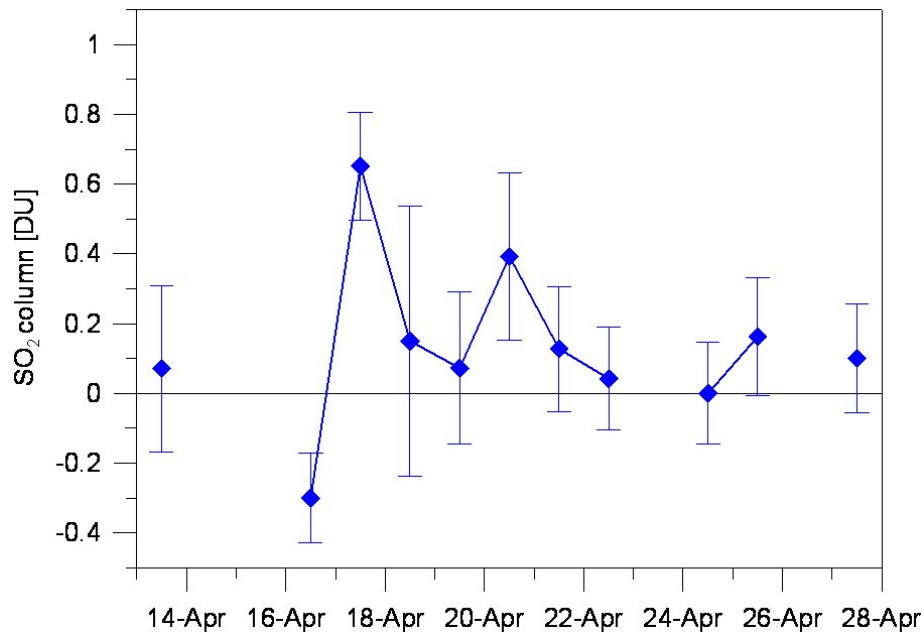
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**Fig. 9.** SO<sub>2</sub> column measurements (daily means) by the Hohenpeissenberg Brewer spectrometer. Maximum column densities of about 0.7 DU were measured on 17 April (corresponding to a 3 km layer with 3 ppb SO<sub>2</sub>).

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