

Monitoring presence and streaming patterns of Icelandic volcanic ash during its arrival to Slovenia

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Abstract. The eruption of the Eyjafjallajökull volcano starting on 14 April 2010 resulted in the spreading of volcanic ash over most parts of Europe. In Slovenia, the presence of volcanic ash was monitored using ground-based in-situ measurements, lidar-based remote sensing and airborne in-situ measurements. Volcanic origin of the detected aerosols was confirmed by subsequent spectral and chemical analysis of the collected samples. The initial arrival of volcanic ash to Slovenia was first detected through the analysis of precipitation, which occurred on 17 April 2010 at 01:00 UTC and confirmed by satellite-based remote sensing. At this time, the presence of low clouds and occasional precipitation prevented ash monitoring using lidar-based remote sensing. The second arrival of volcanic ash on 20 April 2010 was detected by both lidar-based remote sensing and airborne in-situ measurements, revealing two or more elevated atmospheric aerosol layers. The ash was not seen in satellite images due to lower concentrations. The identification of aerosol samples from ground-based and airborne in-situ measurements based on energy-dispersive X-ray spectroscopy confirmed that a fraction of particles were volcanic ash from the Eyjafjallajökull eruption. To explain the history of the air masses bringing volcanic ash to Slovenia, we analyzed airflow trajectories using ECMWF and HYSPLIT models.

1 Introduction

Eyjafjallajökull is one of the smaller ice caps of Iceland, which covers the caldera of a volcano with a summit elevation of 1666 m. Since the eruptions in 1821 to 1823, the Eyjafjallajökull has been inactive for 187 yr (Larsen, 1999). In December 2009, new seismic activity started with thousands of small earthquakes with epicenters beneath the volcano. The first subsequent eruption in the form of a fissure vent began on 20 March 2010. After a brief pause, Eyjafjallajökull resumed erupting on 14 April 2010, throwing volcanic ash approximately 9 km up into the atmosphere. Ash plume led to air travel disruption in northwest Europe for a total of six days starting from 15 April as well as in May 2010, including the closure of airspace over many EU member states (Sanderson, 2010a,b).

Volcanic ash, consisting of small bits of pulverized rock with a diameter of less than 2 mm, is formed during explosive volcanic eruptions. Subsequent ash deposit can eventually lead not only to immediate damage of the local ecosystem, but also to severe disruption of air traffic. Therefore, detecting the concentrations and characteristics of the streaming volcanic ash and thus deducing its impact on local environment is becoming more and more critical in order to minimize possible damage (Witham, 2005; Casadevell, 1994). In previous studies of volcanic ash, in-situ devices and remote sensing techniques were demonstrated to be the most powerful tools for quantitative measurements of its presence (Pieri et al., 2002; Sassen et al., 2007; Delene et al., 1996). Furthermore, in the case of in-situ measurements, the morphology and chemical composition of the volcanic ash can simultaneously be analyzed (Riley et al., 2003; Taylor and Lichte, 1980).



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During and after the eruption of the Eyjafjallajökull volcano, the presence and characteristics of volcanic ash have been monitored over large parts of Europe by both in-situ measurements and remote sensing tools, aiming to minimize the ecological and economic impacts (Sigmundsson and Höskuldsson, 2010; Ansmann et al., 2010; Flentje et al., 2010; Schumann et al., 2011). Slovenia, lying in Central Europe on the crossroad of Mediterranean, Alpine and Continental weather influence, is less than 3000 km away from Iceland and was also affected by the volcanic ash. Its presence was monitored using ground-based in-situ measurements, lidar-based remote sensing and airborne in-situ measurements, which was followed by the subsequent analysis of collected aerosol samples. In this paper, we present the results of the measuring campaign, including the modeling of air-mass trajectories reaching Slovenia, using ECMWF and HYSPLIT models.

2 Synoptic situation over Europe after the eruption

Considering the time of eruption of the Eyjafjallajökull volcano and the distance between Iceland and Slovenia, the synoptic situation over Europe will be focused on the days from 14 to 20 April 2010. At this time, volcanic ash was still largely confined to a localized stream of air masses originating from the Icelandic region and its trajectories could thus be reliably modeled using air-transport models and monitored by satellite imaging.

On 14 April 2010, a high pressure zone was present over the eastern Atlantic Ocean and partially reached the British Islands. The pressure center slowly moved towards central France. At higher altitudes, between 5 km and 10 km over Iceland, the prevailing wind directions were northwesterly, which resulted in a southeast direction of the airflow carrying volcanic ash. Therefore, the volcanic ash plume moved from the North Sea towards Denmark and further toward Central Europe. Based on Meteosat-8 satellite images, air masses containing volcanic ash reached the air space over Slovenia between 00:00–06:00 UTC on 17 April 2010 (Meteosat Data, 2010). Since a high pressure zone dominated most of Europe at this time, the deposition of volcanic ash was only due to gravity, friction and the general descent of air masses.

On 17 April there was a local shower at Ljubljana between 01:00–02:30 UTC. Moreover, a shallow low-pressure center, which formed over Northern Italy and moved towards Slovenia, resulted in further precipitation on 18 April 2010 between 03:20–08:20 UTC at Nova Gorica and 07:00–11:30 UTC at Ljubljana. Precipitation clouds reached altitudes of around 5 km, causing the washout of volcanic ash at lower altitudes. Following the precipitation, winds temporarily changed direction towards the southwest and blew away the remains of volcanic ash. As the low-pressure center moved eastwards, a high-pressure zone over France prevailed, causing the airflow over Slovenia to change direc-



Fig. 1. Map of Slovenia indicating the measuring locations of the campaign. At Nova Gorica and Otlica lidar-based remote sensing were performed. Ground-based in-situ measurements were done in Ljubljana and airborne in-situ measurements were performed during the aircraft ascent at Divača, descent at Vrhnika and during the flight from Divača to Vrhnika (dashed line). The inland map of Europe indicates the position of Slovenia and Iceland.

tion towards the north-northeast. Due to the second change in wind direction, air masses containing volcanic ash could have again arrived over Slovenia, at lower altitudes, on 20 April 2010.

3 Measurements

In order to identify the amount, type and streaming patterns of Icelandic volcanic ash over Slovenia, a measuring campaign including ground-based in-situ measurements, lidar-based remote sensing and airborne in-situ measurements was performed. Not all the measurements were performed during the entire measuring campaign, as this was not permitted by the nature of the experimental setup as well as the local weather conditions. In addition, the aircraft-based measurements were only possible along a specific flight route. The locations of the measurement sites are shown in Fig. 1 and the distances between them are listed in Table 1. All the sites are within a circular zone with an approximate radius of 35 km.

3.1 Ground-based measurements

Concentrations of solid particles (PM_{10}) and sulphur dioxide (SO_2) in the air along with the chemical composition of precipitation are monitored routinely by Slovenian Environment Agency in Ljubljana. The measurement site is located in the city center, which may be influenced by traffic and heating effects. In order to determine whether volcanic ash that was expected to be streaming above Slovenia had been deposited and had any impact on local environment, the collected data for the period from 10 April to 3 May 2010 has been studied in detail, starting well before the Eyjafjallajökull eruption

Table 1. Distance between measurement sites used in the campaign.

Distance [km]	Nova Gorica	Otlica	Divača	Vrhnika	Lat., Lon.
Nova Gorica					45.96° N, 13.64° E
Otlica	20.5				45.93° N, 13.91° E
Divača	39.2	28.0			45.68° N, 13.97° E
Vrhnika	50.5	30.5	40.6		45.97° N, 14.29° E
Ljubljana	67.5	48.0	58.5	18.6	46.05° N, 14.50° E

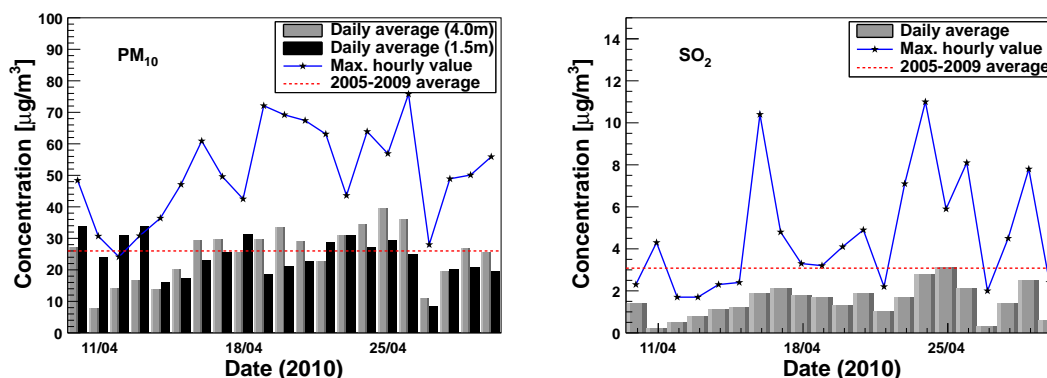


Fig. 2. Monitoring results of the concentrations of PM₁₀ at 1.5 m and 4.0 m (left) and SO₂ at 4.0 m (right) above the ground in Ljubljana from 10 April to the end of April 2010. The maximum hourly value of PM₁₀ represents the larger value obtained at the two measuring heights.

and ending after the volcanic ash had spread all over Europe, including Slovenia.

Concentration of the PM₁₀ particles was continuously monitored at two different heights (1.5 m and 4.0 m above the ground) using a tapered element oscillating microbalance¹. The daily average values at these two heights and the maximum hourly value are shown in Fig. 2 (left). Although a slight increase in the 4.0 m data can be seen between 16–26 April and the maximum hourly concentration of PM₁₀ exceeded 60 µg m⁻³ on certain days (16, 19–22, 24 and 26 April), most daily averages are lower or comparable to the long term average between 2005–2009. Concentrations of SO₂ were continuously monitored at 4.0 m above the ground using an ultraviolet fluorescent SO₂ analyzer². The daily average data and the maximum hourly value are shown in Fig. 2 (right). The average concentration of SO₂ increased linearly from 11 to 17 April, then decreased linearly until 22 April with the exception of a single peak on 21 April. Similar to PM₁₀ data, most values are lower than the long term average between 2005–2009. As the concentrations of PM₁₀ and SO₂ never significantly exceeded the long term average values, these measurements were used only to show the extent of the environmental impact of the deposition of volcanic ash in Slovenia, which was found to be very limited.

¹TEOM series 1400a ambient particulate monitor, Thermo Electron Corporation, USA

²Model 8850, Teledyne Monitor Labs Inc., USA

Monitoring of ash deposition and concentration in the precipitation depended on its occurrence, which was predominantly between 10–14 April, 17–19 April, 23 April and 2–3 May. On 10, 19 and 23 April, the precipitation was very light and did not yield a sample large enough to allow chemical analysis. Concentration of anions (F⁻, Cl⁻, NO₃⁻ and SO₄²⁻) in the samples was checked by ion chromatography using an analytical column³ together with a conductivity detector. Results of the analysis are shown in Fig. 3. Concentrations of selected metals (Cr, Ni, Cs, Pb, Cu and As) were measured in acidified samples using a mass spectrometer⁴. Among these, only the presence of F⁻, which is usually below the detection limit, was an indication of the presence of aerosols from volcanic activities (Oppenheimer et al., 2003). F⁻ anions were observed on 13, 14, 17 and 18 April and 2 May, with highest concentration of 0.45 mg l⁻¹ on 17 April and highest deposition of 6.64 mg m⁻² day⁻¹ on 18 April 2010. Small traces of F⁻ on 13 and 14 April (about 10 times smaller than on 17 April) may be due to the ashes from the first phase of smaller eruptions of Eyjafjallajökull starting from 20 March 2010. A fraction of F⁻ anions remained present in precipitation as long as 2 May 2010. A possible explanation is that air masses with a certain amount of volcanic-ash content kept streaming over and around Slovenia, and the precipitation on 2 May 2010 washed out all the remaining volcanic

³IonPac AS14 analytical column, Dionex Corporation, USA

⁴ICP-MS, Thermo Electron Corporation, USA

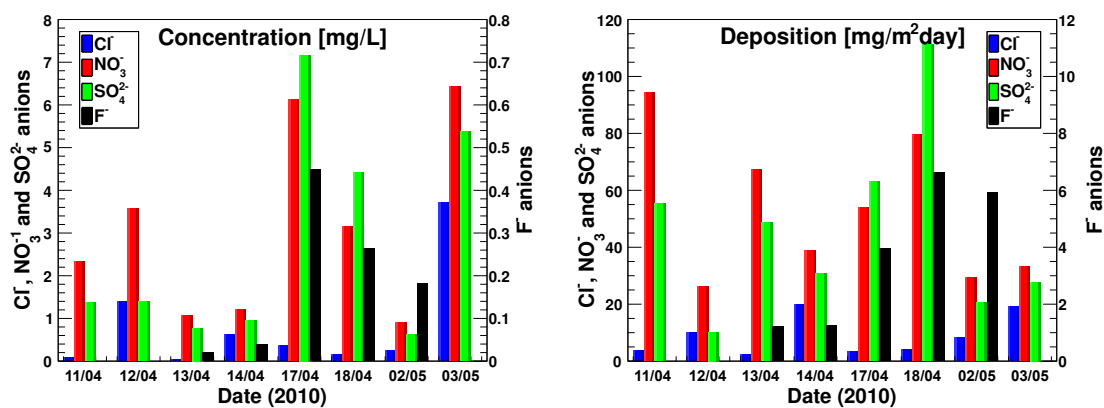


Fig. 3. Results of the composition analysis of precipitation in Ljubljana between 10 April and 3 May 2010, showing concentrations (left) and depositions (right) of various anions (Cl^- , NO_3^- , SO_4^{2-} and F^-) in the precipitation.

ash particles. F^- anions were no longer present in the precipitation on 3 May 2010.

In parallel to the monitoring of particle concentrations, particle sampling was also performed from 10 to 18 April 2010 using a sequential particle sampler with PM_{10} and $\text{PM}_{2.5}$ reference inlets. Spectral and chemical analysis of particle samples showed the presence of volcanic ash on 17 and 18 April 2010. Detailed analysis is presented in Sect. 4.

3.2 Lidar-based remote sensing

During the days when air masses might have carried volcanic ash to Slovenia, lidar-based remote sensing of the atmosphere was performed by University of Nova Gorica to probe for its optical characteristics. It was based on vertical measurements of two non-coaxial Mie scattering lidar systems located at Nova Gorica (107 m a.s.l.⁵) (He et al., 2010) and at Otlica (945 m a.s.l.) (Gao et al., 2011). Specifications of both lidar systems are summarized in Table 2. In both cases, data acquisition was performed using transient recorders⁶ in combination with C++ (Linux) based data acquisition software. Default range resolution of the digitizer is 3.75 m with a trace length of 16k. In the data processing of both lidar systems, 3 consecutive bins were merged into one bin to improve the signal-to-noise ratio, thus reducing the range resolution to 11.25 m.

Measurements using the Nova Gorica lidar were performed consecutively for four days (17 to 20 April 2010), while the measurements using the Otlica lidar were performed only on 20 April 2010 (20:00–03:00+1 CET⁷). On 17 and 18 April, low clouds and occasional precipitation limited the lidar detectable range to about 2 km, thus preventing aerosol monitoring at higher altitudes. On 19 April, the

⁵a.s.l. is an abbreviation for “above sea level”

⁶TR40-160 transient recorder, Licel, Germany

⁷CET – Central Europe Time, 1 h ahead of Coordinated Universal Time (UTC)

Table 2. Specifications of the non-coaxial Mie scattering lidar systems.

Site	Nova Gorica	Otlica
Transmitter	Big Sky CFR400	Quantel Brilliant B
Wavelength	1064 nm	355 nm
Pulse energy	40 mJ	95 mJ
Repetition rate	10 Hz	20 Hz
Receiver	Newtonian telescope	Parabolic mirror
Diameter	300 mm	800 mm
Focal length	1500 mm	410 mm
Detector	EG&G APD	Hamamatsu PMT
Type	C30954	R7400
Voltage	300 V	770 V
Complete overlap	> 400 m	> 1000 m

sky was clear but no sharp peaks in the lidar return signal were detected. On 20 April, the sky was also clear, except for some middle and high altitude clouds which appeared in the evening and limited the detectable range to about 5 km. On this day, one or more elevated aerosol layers were observed by both lidar systems in each of the performed measurements (10:10–11:10, 15:20–17:50 and 19:40–22:50 CET at Nova Gorica and 20:00–01:00+1 CET at Otlica). Figure 4 provides an example of the Nova Gorica lidar observations performed from 20:00 to 22:50 CET on 20 April 2010. Two elevated aerosol layers can be clearly seen. The upper one was predominantly at the altitude of 2.5–3.0 km, while the altitude of the lower one was increasing with time from 1.0 to 1.5 km. Similar trends were found in Otlica data. Throughout the lidar operation, meteorological data (temperature, humidity, air pressure and wind velocity) were also monitored by the weather stations co-located at both lidar sites. Meteorological conditions on 20 April were quite similar to that of 19 April as well as to clear sky conditions before

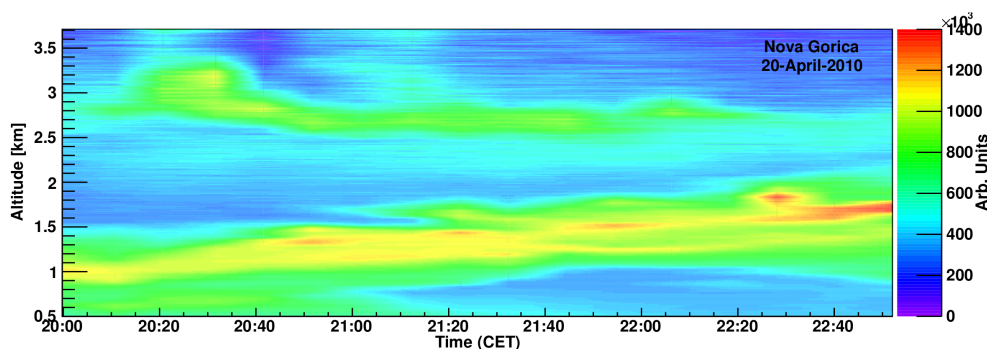


Fig. 4. Temporal variation of aerosol layers over Nova Gorica between 20:00–22:50 CET on 20 April 2010 in terms of 1064-nm range-corrected lidar signal (arbitrary units). Two elevated aerosol layers can be clearly seen during the measurement, which are at altitudes of 1.0–1.5 km and 2.5–3.0 km, respectively.

the Eyjafjallajökull volcano eruption, implying that the local phenomena directly above the ground on 20 April 2010 could have had only limited impact on the observed vertical atmospheric structure.

In the case of both lidar systems, the aerosol optical variable (backscatter coefficient) was calculated using the Klett method (Klett, 1981) to make the retrievals comparable. In the calculation, the lidar ratio was set to 55 ± 5 sr, according to the extinction-to-backscatter ratios of the ash plume measured by the EARLINET lidars (Ansmann et al., 2010), and the reference height was set to the height where just pure molecular attenuation took place (about 5–6 km a.s.l. for both Nova Gorica and Otlica). The molecular contribution estimated by the US Standard Atmosphere model (US Standard Atmosphere, 1976) was subtracted after the inversion procedure. The calculation results for each consecutive measurement interval are shown in Fig. 5.

From the Nova Gorica data, the presence of an upper elevated aerosol layer at the altitude of about 3 km can be seen throughout the day, while the lower elevated aerosol layer at the altitude of about 1.3 km appeared in the evening measurements. Temporal evolution of these two layers (peak backscatter coefficients of the two layers and their corresponding altitudes) are presented in Fig. 6. The altitude of the upper aerosol layer slowly decreased from 3.15 km in the morning to 2.77 km in the evening, after atmospheric motion subsided. The corresponding average peak value of the aerosol backscatter coefficient was found to be smaller in the afternoon ($0.439(1 \pm 0.66) \times 10^{-3} \text{ km}^{-1} \text{ sr}^{-1}$) than in the morning ($0.725(1 \pm 0.35) \times 10^{-3} \text{ km}^{-1} \text{ sr}^{-1}$) and in the evening ($0.581(1 \pm 0.28) \times 10^{-3} \text{ km}^{-1} \text{ sr}^{-1}$), implying that the aerosol concentration was decreasing in the morning and then increasing again in the afternoon. Large uncertainties in the afternoon data imply that atmospheric conditions were most turbulent in the afternoon and that atmospheric motion was strong during the measurement period. The lower aerosol layer initially appeared in the measurement profile at 16:50 CET, and its altitude increased linearly

with time from 0.6 km (lowest value in the afternoon) to 1.75 km (highest value in the evening). In the evening, the average peak value of the backscatter coefficient in this layer was $0.712(1 \pm 0.16) \times 10^{-3} \text{ km}^{-1} \text{ sr}^{-1}$, even higher than the upper layer.

In the Otlica data from 20:00 CET, all the profiles show the presence of two elevated aerosol layers at about 2.6 km and 1.7 km. Temporal evolution of the altitude of the aerosol backscatter peaks and magnitudes of the peak backscatter coefficients are shown in Fig. 7. The altitude of the upper aerosol layer varied between 2.35 km and 2.65 km with an average value of 2.51 km. The corresponding backscatter coefficient was found to be almost constant in certain time intervals (20:20–21:20, 21:40–23:10 and 00:00–01:00 CET of the next day). Its value increased from $0.579 \times 10^{-3} \text{ km}^{-1} \text{ sr}^{-1}$ at around 20:00 CET to $2.88 \times 10^{-3} \text{ km}^{-1} \text{ sr}^{-1}$ at around 01:00+1 CET, implying that the aerosol loading gradually increased during the night. The lower aerosol layer was initially observed at 1.78 km. Its altitude linearly decreased to 1.60 km at 21:00 CET, then increased again to about 1.95 km at 00:50+1 CET. The average peak altitude of this layer was 1.77 km. Even though the lower aerosol layer was not within the complete overlap region of the Otlica lidar system, the peak value of its backscatter coefficient exceeded the one from the upper layer after 23:00 CET, which implies that at that time aerosol loading was greater in the lower layer.

The comparison of aerosol optical variables simultaneously obtained using Nova Gorica and Otlica lidars (20:00–22:50 CET on 20 April 2010) also revealed, in addition to the agreement in the elevation of the altitude of the upper aerosol layer, a spectral dependence of aerosol extinction (α) within this layer,

$$\frac{\alpha_{\text{Otlica}}}{\alpha_{\text{NovaGorica}}} = \left(\frac{\lambda_{\text{Otlica}}}{\lambda_{\text{NovaGorica}}} \right)^{-m} \simeq 3,$$

As we assumed a fixed lidar ratio (S) in all lidar measurements, aerosol extinction and backscatter coefficient (β) are correlated as $\alpha = S \cdot \beta$. The Angström exponent (m) was

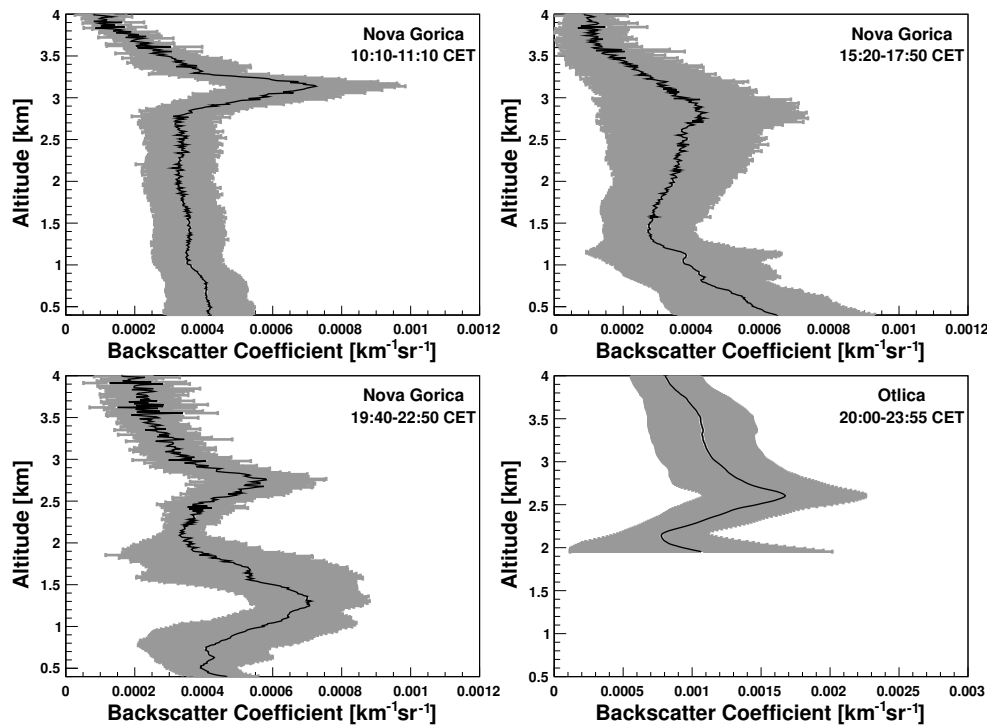


Fig. 5. Aerosol backscatter coefficients calculated by the Klett method during each time interval of the lidar measurements. The black line represents the mean value during each interval and the grey shaded areas the corresponding uncertainties of the measurement. Only the data within complete overlap is presented.

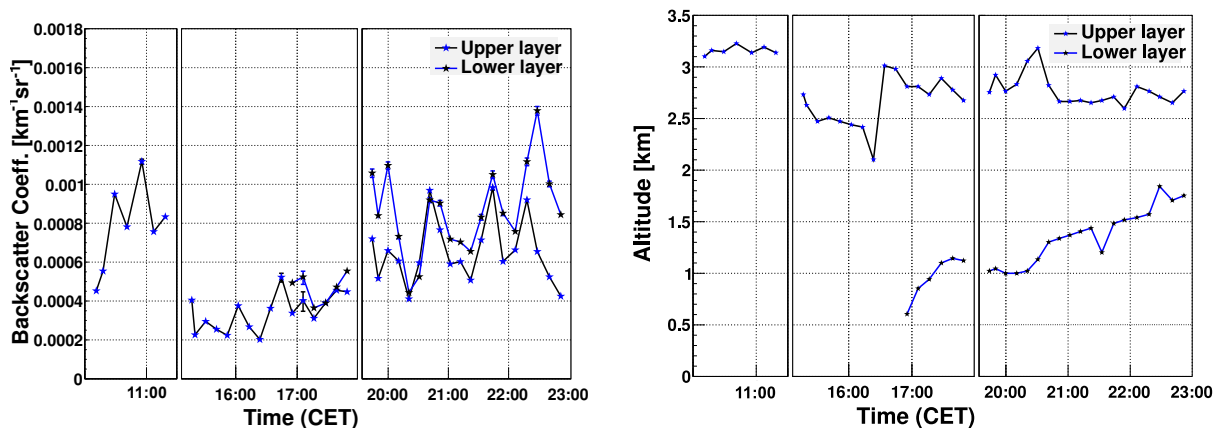


Fig. 6. Variation of peak aerosol backscatter coefficients for the two observed aerosol layers (left) and their corresponding altitudes (right) measured by the Nova Gorica lidar on 20 April 2010. Each data point represents an observation with 10 min integration time (6000 laser shots). For the backscatter coefficient, errors bars denote the uncertainty due to the set lidar ratio.

found to be close to one, which implies the average particle size in the upper aerosol layer was small (Russell et al., 2010). This was also confirmed by airborne particle sampling described in the next section. The lower aerosol layer cannot, however, be compared between the two sites due to the possibility of site-specific mixing of volcanic ash with local aerosols and the fact that at Otlica the lower layer was not within the complete overlap region of the system.

3.3 Airborne measurements

On 20 April 2010 (15:00–15:38 CET), an airborne mission aiming at sampling the ash particles and measuring their concentration was carried out by Istituto Nazionale di Oceanografia e di Geofisica Sperimentale using a Cessna 172 aircraft. The aircraft performed a spiral climb from 0.61 to 3.05 km at Divača (15:00–15:16 CET), followed by a leveled

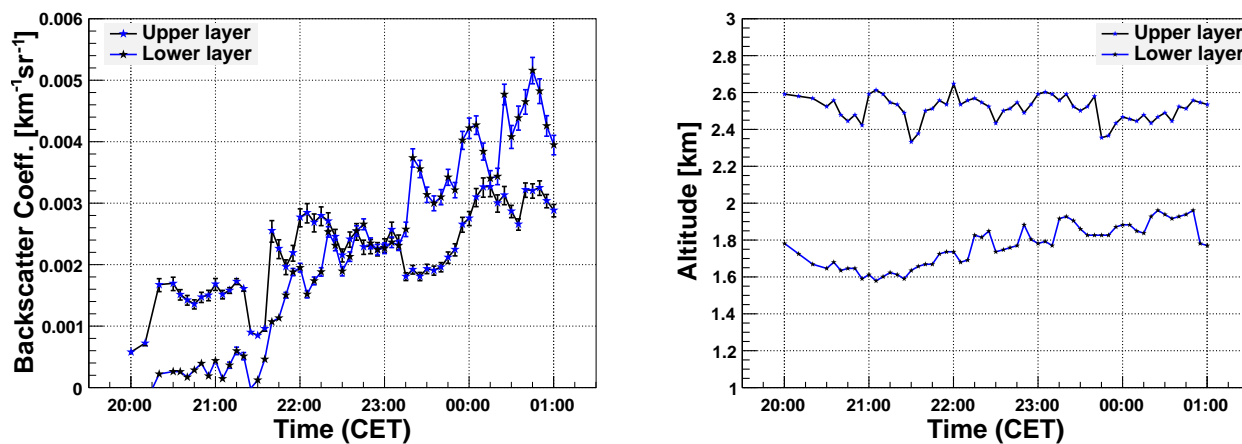


Fig. 7. Variation of peak aerosol backscatter coefficients for the two observed aerosol layers (left) and their corresponding altitudes (right) measured by the Otlica lidar during the night of 20 April 2010. Each data point represents an observation with 5 min integration time (6000 laser shots). For the backscatter coefficient, error bars denote the uncertainty due to the set lidar ratio.

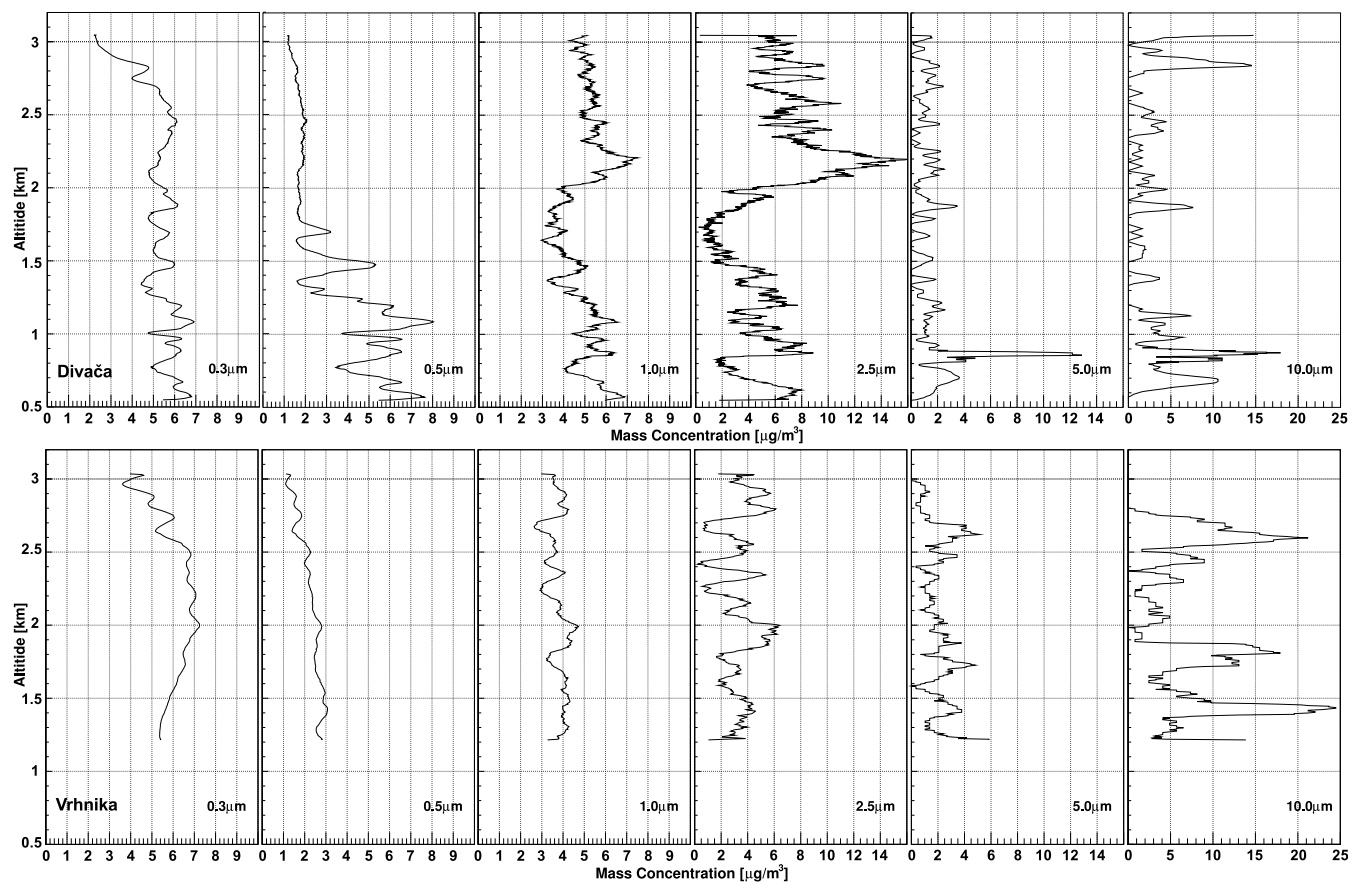


Fig. 8. Aerosol concentration profiles for six different aerosol dynamic equivalent diameters measured on 20 April 2010 above Divača (top) and above Vrhnika (bottom).

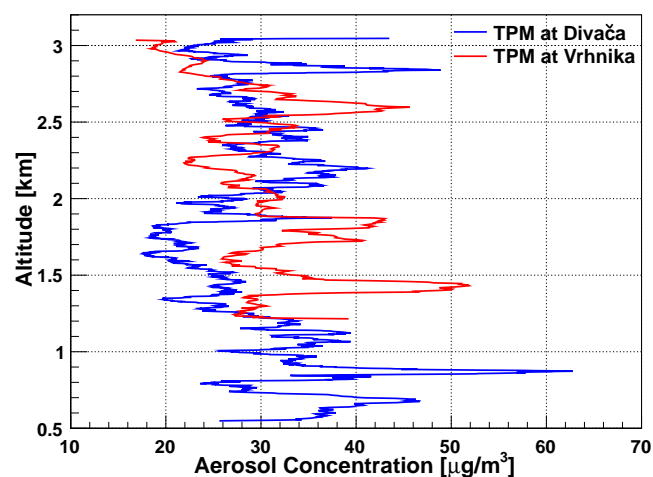


Fig. 9. Profiles of the total particle matter content in the atmosphere above Divača (blue) and above Vrhnika (red) on 20 April 2010 as obtained from the laser particle counter.

flight from Divača to Vrhnika (15:16–15:26 CET) and a spiral descent at Vrhnika to 1.22 km (15:26–15:38 CET) with a constant rate of ascend and descend of 2.5 m s^{-1} . Devices used for sampling aerosol particles and for measuring aerosol concentration were mounted inside the aircraft. Outer air from the intake at the aircraft wing was split into two parts. One half was conducted into an expansion box, where it was strongly decelerated and subsequently used as the input to a laser particle counter⁸. The counter simultaneously provided particle concentrations for six different aerodynamic equivalent diameters (AED) of 0.3, 0.5, 1.0, 2.5, 5.0 and $10.0 \mu\text{m}$. Its sampling rate was set to 2 s, with the corresponding step in elevation of 5 m at the adopted ascend/descend rate.

As the laser particle counter can only provide particle density for any given AED, the assumptions on particle shape (spherical) and constant density of the particulate matter (2500 kg m^{-3}) were made to obtain the aerosol mass concentration. Mass concentration profiles for six different AED categories measured above Divača and above Vrhnika are shown in Fig. 8. Above Divača, aerosol concentration for $0.3 \mu\text{m}$ and $0.5 \mu\text{m}$ particles manifests similar features, such as “ripples” below 2 km and clearly visible layers at 1.1 km and 1.5 km. Concentration profiles of $1.0 \mu\text{m}$ and $2.5 \mu\text{m}$ particles show a distinct aerosol layer at 2.2 km. Below 2 km, the profile of $2.5 \mu\text{m}$ particles shows the presence of three additional layers (at 0.6 km, 0.9 km and 1.2 km) with particle concentrations exceeding $7.0 \mu\text{g m}^{-3}$ in each layer. Concentration profiles of $5.0 \mu\text{m}$ particles manifests more “ripples” except for a clearly visible peak at 0.85 km with a magnitude of $13.0 \mu\text{g m}^{-3}$. The peak at this altitude can also be seen in the distribution of $10.0 \mu\text{m}$ particles with similar magni-

tude. Moreover, distribution of $10.0 \mu\text{m}$ particles shows other peaks at 1.15 km, 1.85 km, 2.4 km and 2.8 km. Compared to Divača, the distributions of $0.3 \mu\text{m}$, $0.5 \mu\text{m}$ and $1.0 \mu\text{m}$ particles were much smoother above Vrhnika, showing only a small peak at 2 km. Distribution of $2.5 \mu\text{m}$ particles has more layers, the most prominent ones being those at 1.9 km and 2.8 km. The concentration for $5.0 \mu\text{m}$ particles decreased between the altitudes of 1.7 km and 2.3 km, then increased up to 2.7 km, but the observed variations around the mean ($3.5 \mu\text{g m}^{-3}$) were rather small. However, from the distribution of $10.0 \mu\text{m}$ particles three aerosol layers (at 1.45 km, 2.4 km and 2.6 km) can clearly be seen. Peak values of the aerosol concentration in all these layers exceed $17.0 \mu\text{g m}^{-3}$. Distributions of the total particle matter (TPM) above Divača and above Vrhnika including all the above AED categories are plotted in Fig. 9. Except for the peak at the height of 0.85 km which may be within the atmospheric boundary layer and thus influenced by local activities (Garratt, 1992), peaks with the maximum aerosol concentration exceeding $40 \sim 50 \mu\text{g m}^{-3}$ were found at the altitudes of 2.2 km and 2.8 km above Divača and at the altitudes of 1.45 km, 1.85 km and 2.6 km above Vrhnika.

The other half of the unfiltered conveyed air was manually directed to impinge upon standard double-sided adhesive tape pads, which were replaced at regular time intervals. The sampling procedure yielded 9 samples during the ascent of the aircraft which represent the cumulative effect of aerosols in the elevation intervals of 550–640 m, 850–940 m, ... and 2950–3040 m. Three additional adhesive tapes were fixed on the aircraft wing struts to sample the aerosols in the entire air column. Detailed spectral and chemical analysis of the particle samples collected during the flight are described in the following section.

4 Identification of ash particles

Identification of volcanic ash in particle samples was based on particle-by-particle comparison of their chemical and petrographic composition to that of the reference volcanic ash sample collected on 24 July 2010 in the southern part of Iceland, about 50 km away from the Eyjafjallajökull volcano after the main eruption. Volcanic ash candidates from all the samples (ground-based measurements and airborne measurement) were analyzed using a low-vacuum scanning electron microscope (SEM) using backscattered electron image mode and energy dispersive X-ray micro-analysis⁹. Rejected particles were either minerals or solids such as chert, gypsum, limestone or salt, or aerosols of biological origin such as pollen. Structurally, volcanic ash was found to appear in two forms: either as single, angular-shaped particles or as agglomerates with sizes up to a few tens of microns. An agglomerate was found to be comprised of a large number of heterogeneous grains typically up to a micron in size.

⁸Lighthouse 3016 IAQ Particle Counter, Lighthouse Worldwide Solutions, USA

⁹JEOL 5500 LV, JEOL Ltd., Japan

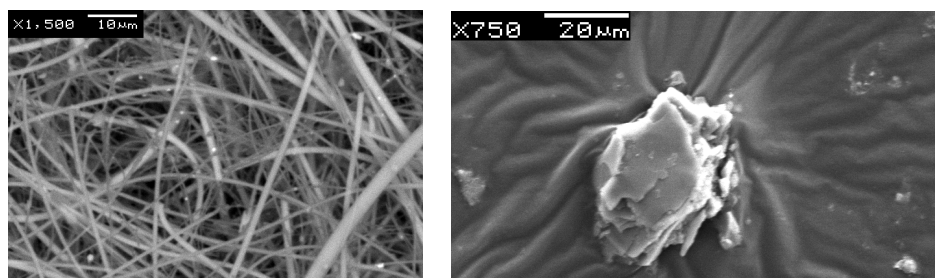


Fig. 10. SEM images of aerosol particles obtained from ground-based and airborne measurements. Left: image of a filter used in the sequential sampler on 17 April 2010 (1500-fold magnification). Bright points on the filter were found mostly to be ash particles. Right: image of an agglomerate ash particle collected at the altitude of 2.2 km a.s.l. (750-fold magnification).

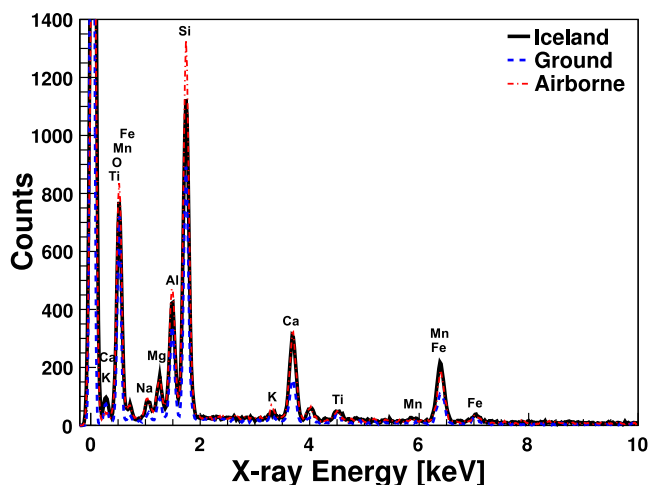


Fig. 11. Energy dispersive X-ray spectrum of the reference ash sample obtained from Iceland (full black line) and the ash sample collected in the ground-based (dashed blue line) and airborne (dotted red line) measurements in Slovenia. The reference sample was collected in the southern part of Iceland, about 50 km away from the Eyjafjallajökull volcano, after the main eruption.

The comparison of the energy dispersive X-ray spectra of the reference ash sample and the samples collected in Slovenia shows good agreement between the three samples (Fig. 11). Ash particle candidates from Slovenia contained the representative elements (Si, Al, K, Mg, Ca, Fe and Ti) in ratios comparable to the values found in the reference ash samples. Detailed chemical composition of ash candidates and ash from the reference samples are presented in Table 3. The only notable difference between the two is a trace of MnO in the reference sample, which was not observed in any of the particle samples collected in Slovenia.

In particle samples from the ground-based measurements, ash candidates were found on 17 and 18 April 2010, predominantly in the form of small-size grains (about 1 μm) and a small number of agglomerated particles with sizes up to 20 μm . In the SEM image of the filter of the sequential sam-

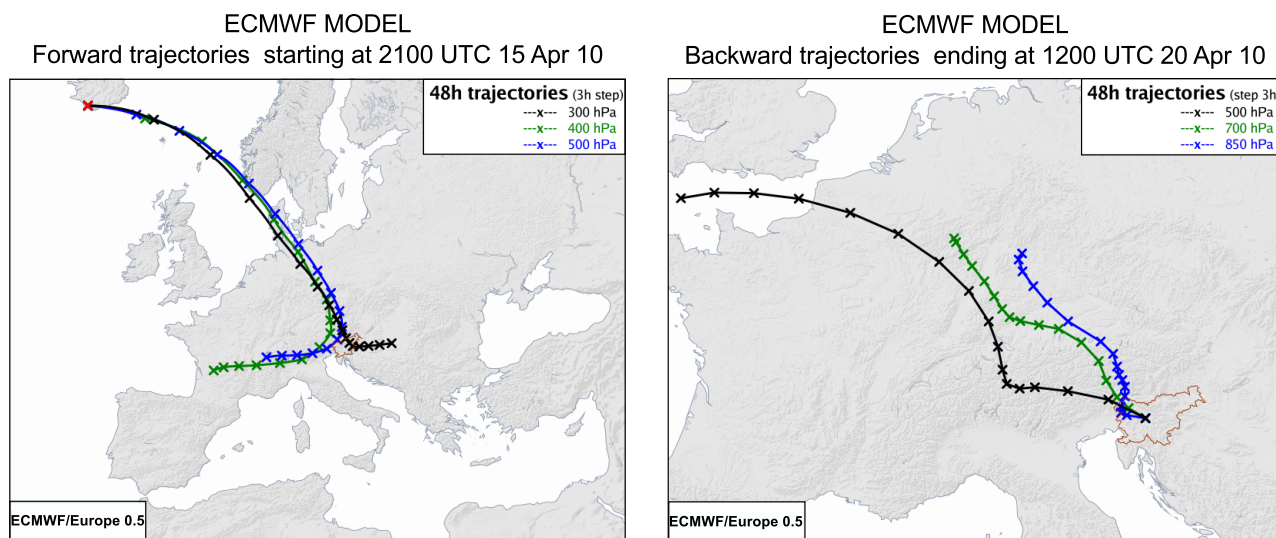
pler device (Fig. 10, left), the collected ash particle candidates can be seen as bright dots. The volume fraction of volcanic ash with respect to all the collected particles was found to be 30 % on 17 April and 15 % on 18 April 2010. In particle samples from the airborne measurement (20 April 2010) ash candidates were found at all nine sampling altitudes with a peak surface density of approximately 36 particles mm^{-2} (47 % of all particles) at 2.2 km a.s.l. Ash particle candidates were found to be predominantly agglomerated particles (Fig. 10, right) with sizes up to a maximum of 70 μm , which were, for the purpose of determining ash particle surface density on the adhesive tape, regarded as a single particle rather than composites of heterogeneous grains.

5 Simulation of air flow trajectories

Results of chemical analysis of the aerosol samples indicate that the volcanic ash reached Slovenia at least twice: on 17 and 20 April. To determine the paths of its transport from Iceland and the time when it was emitted into the atmosphere, we simulated the transporting air flow trajectories using the HYSPLIT model (HYSPLIT Data, 2010) and the ECMWF model (ECMWF Data, 2010). The simulation of forward trajectories originating over Iceland (63.63° N, 19.62° W), performed for the period of 14–19 April with one hour increment of the starting time, established that trajectories starting before 15 April 2010, 21:00 UTC do not overpass Slovenia. Both models predict that air masses carrying volcanic ash started from Eyjafjallajökull region between 21:00 UTC and 04:00+1 UTC on 15 April 2010 and reached Slovenia from the north at relatively high altitudes (> 5 km) between 01:00–02:30 UTC on 17 April 2010 (Fig. 12, left and Fig. 13, left). In the output of the ECMWF model, the heights of airflow trajectories are shown in standard atmospheric pressures of 300 hPa, 400 hPa and 500 hPa, corresponding to the altitudes of 9.2 km, 7.2 km and 5.6 km, respectively. In the HYSPLIT model, starting altitudes of 9.3 km, 7.2 km and 5.7 km were selected to make the forward simulation of airflow trajectories. In both simulations, air-mass trajectories at lower altitudes turned towards the west

Table 3. Comparison of the chemical composition between representative particles from the reference ash samples collected in Iceland and ash candidates from the aerosol samples collected in Slovenia.

	Al ₂ O ₃	SiO ₂	K ₂ O	Na ₂ O	CaO	TiO ₂	MgO	FeO	MnO
Iceland	14.4 %	48.6 %	0.7 %	2.8 %	10.3 %	1.9 %	4.8 %	15.7 %	0.8 %
Ground	15.3 %	52.6 %	1.2 %	2.5 %	9.7 %	1.6 %	4.4 %	12.7 %	/
Airborne	16.3 %	53.7 %	0.9 %	1.6 %	7.6 %	2.9 %	3.6 %	13.4 %	/

**Fig. 12.** Air flow trajectories for a 48-h time interval with 3-h temporal resolution obtained from the ECMWF model. Left: forward trajectories for air masses originating near the Eyjafjallajökull volcano starting on 15 April 2010 at 21:00 UTC; Right: backward trajectories of air masses reaching Ljubljana on 20 April 2010 at 12:00 UTC.

and passed through northern Italy, Switzerland and central France, while the one at the highest-altitude turned towards the east and passed through northern Croatia and Hungary.

For the second arrival of volcanic ash on 20 April, 48-h forward trajectories show that ash-loaded air masses did not arrive directly from Eyjafjallajökull, so 48-h backward trajectories were checked as well by both models for the end-time period from 16–21 April. In the case of ECMWF, Ljubljana was the single endpoint and in the case of HYSPLIT multiple endpoints included Ljubljana, Nova Gorica and Divača. Simulations show that volcanic ash arriving on 20 April was brought by air masses continuously streaming for more than 15 h (starting after 19 April 2010 at 23:00 UTC) from northern and north-western Europe, where ash particles were still present (Flentje et al., 2010; Schumann et al., 2011). The streaming altitudes were found to be below 6 km. Examples of backward airflow trajectories ending at 12:00 UTC on 20 April 2010 are presented in Fig. 12 (right) and Fig. 13 (right). In the ECMWF model, the altitudes of airflow trajectories are presented in standard atmospheric pressures of 500 hPa, 700 hPa and 850 hPa, corresponding to the altitudes of 5.6 km, 3.0 km and 1.5 km, respectively. In the HYSPLIT

model, the endpoint altitudes of 5.6 km, 3.0 km and 1.5 km were selected to make a comparable backward simulation.

6 Discussions and conclusions

The first arrival of volcanic ash to Slovenia was observed in satellite images by Meteosat-8 on 17 April 2010 (Meteosat Data, 2010). At this time due to local weather conditions and high streaming altitudes, the ash was detected through the analysis of the precipitation and particle samples obtained at the ground level. Based on the results of the presented monitoring campaign and the simulation of airflow trajectories, we claim that the volcanic ash from Eyjafjallajökull eruption, which initially reached Slovenia on 17 April 2010, returned at lower altitudes on 20 April 2010 due to the changing of weather conditions over Europe, especially in the Alpine region. The second arrival of volcanic ash was observed by lidar-based remote sensing and airborne measurements, but could not be seen in Meteosat-8 satellite images (Meteosat Data, 2010). Additionally, multi-spectral Meteosat-9 data from the Spinning Enhanced Visible and Infrared Imager with a refresh time of 15 min was used to check for

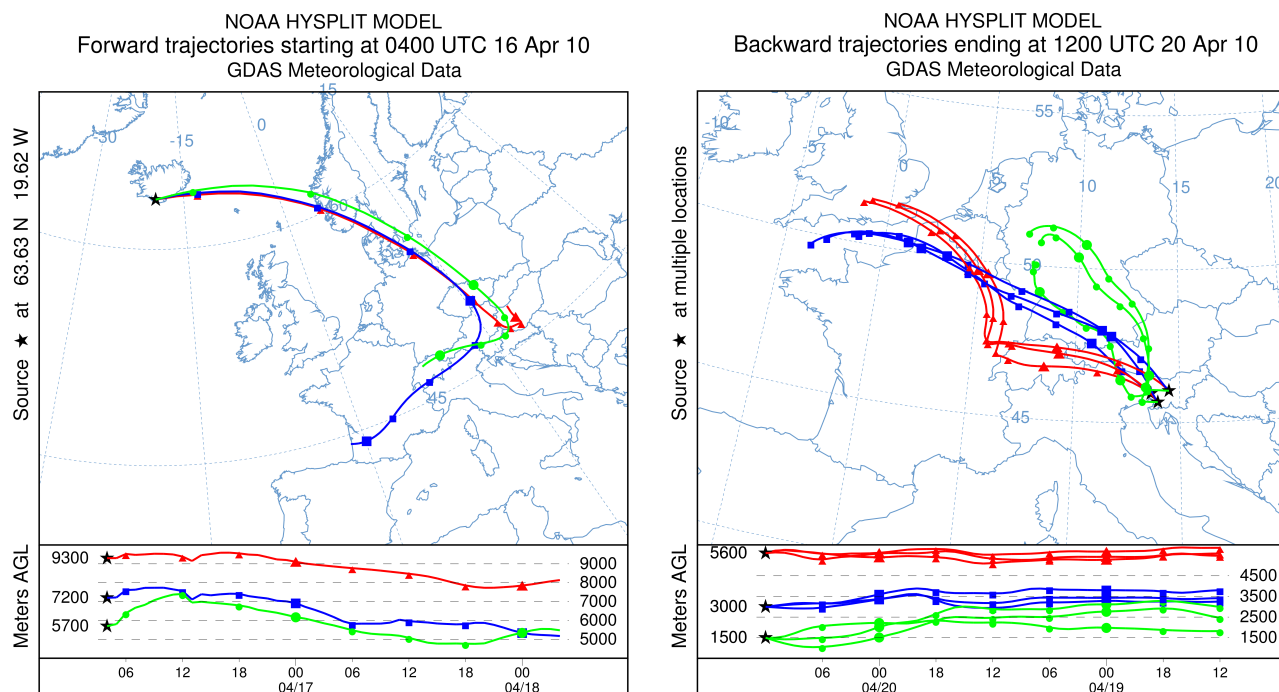


Fig. 13. Air flow trajectories for a 48-h time interval with 6-h temporal resolution obtained from the HYSPLIT model. Left: forward trajectories for air masses originating near the Eyjafjallajökull volcano starting on 16 April 2010 at 04:00 UTC; Right: backward trajectories of air masses reaching Ljubljana, Divača and Nova Gorica on 20 April 2010 at 12:00 UTC.

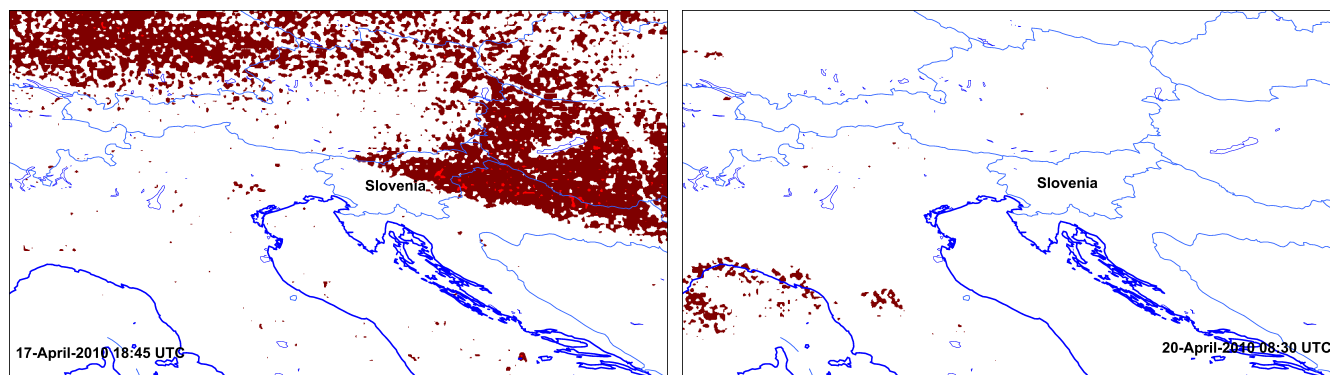


Fig. 14. Meteosat-9 satellite data from Spinning Enhanced Visible and Infrared Imagery show the presence of volcanic ash over Slovenia in detectable quantities on 17 April (left) and its absence on 20 April 2010 (right). Dark red areas indicate the brightness temperature difference below -0.5 K and red areas below -1 K from channels centered at $10.8 \mu\text{m}$ and $12 \mu\text{m}$.

the presence of volcanic ash. The results of the analysis, which combined the channels centered at $8.7 \mu\text{m}$, $10.8 \mu\text{m}$ and $12 \mu\text{m}$ into dust RGB (Lensky et al., 2008) and the difference between brightness temperatures from channels centered at $10.8 \mu\text{m}$ and $12 \mu\text{m}$, unmistakably confirmed ash clouds over Slovenia on 17 April 2010 (throughout the day), but there was no observable signal on 20 April (Fig. 14).

Based on the results of lidar-based remote sensing, we can conclude that the same type of scatters were detected in the elevated aerosol layer at the altitude of around 3 km by

two lidar systems, located about 20 km apart. Airborne measurements yielded peak values for the TPM content of about $40 \sim 50 \mu\text{g m}^{-3}$ during both the aircraft ascent and the 40 km separated descent. Based on the petrographic and chemical identification of collected particle samples, we concluded that aerosol layers detected by both lidar systems were, in fact, layers containing volcanic ash particles. Furthermore, due to the proximity of lidar sites to airborne measurement locations, we assumed it was reasonable to establish a relationship between the aerosol backscatter coefficient (β) and

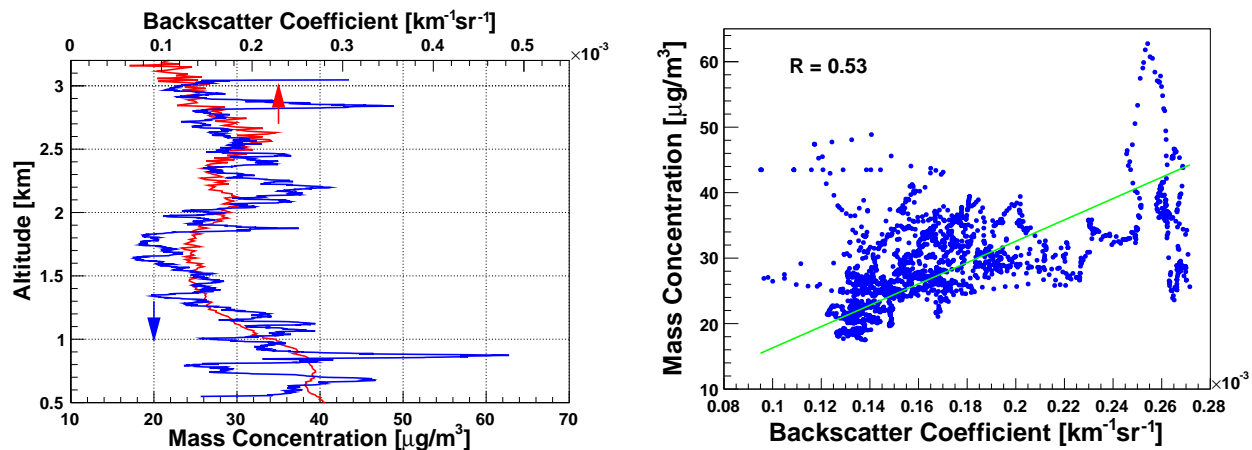


Fig. 15. Total particle matter (TPM) content from the airborne measurement and aerosol backscatter coefficient obtained by lidar were found to be correlated. The correlation can be seen qualitatively from the overlap of the TPM (c) profile over Divača and aerosol backscatter coefficient (β) profile over Nova Gorica simultaneously measured at 15:20 CET on 20 April 2010 (left). The correlation between c and β was found to be linear, $c = (162914 \pm 765)\beta$, with a correlation coefficient of 0.53 (right). In the fit, β was constrained to zero for the limit case when the TPM is zero.

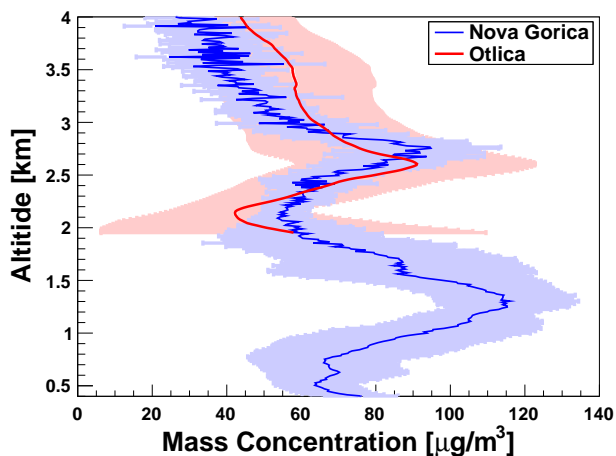


Fig. 16. Mass concentrations of volcanic ash content in the atmosphere above Nova Gorica and Otlica during the first half of the night on 20 April estimated from the approximate 4-h average of the corresponding aerosol backscatter coefficient profiles. Shaded areas denote the uncertainties. In both cases, only the data within the complete overlap is presented.

the TPM content (c) (Wilks, 1995). Using simultaneous measurements of TPM over Divača and aerosol backscatter coefficient over Nova Gorica, the correlation between c and β_{1064} was found to be linear, $c = k_{1064} \cdot \beta_{1064}$, with $k_{1064} = 16.2914(1 \pm 0.005) \text{ km sr } \mu\text{g m}^{-3}$ and a correlation coefficient of 0.53 (Fig. 15). In the fit, β_{1064} was constrained to zero for the limiting case when the TPM is zero. A step of $0.1 \times 10^{-3} \text{ km}^{-1} \text{ sr}^{-1}$ in aerosol backscatter coefficient for 1064 nm laser thus corresponds to aerosol concentration of about $16.3 \mu\text{g m}^{-3}$. According to spectral depen-

dence of aerosol extinction in the upper layer, the correlation between the aerosol backscatter coefficient for 355 nm laser and aerosol concentration was taken to be $k_{355} \simeq k_{1064}/3$. The approximate 4-h average concentrations of volcanic ash above Nova Gorica and Otlica during the first half of the night on 20 April 2010 obtained on the basis of this assumption are shown in Fig. 16, where shaded areas denote statistical uncertainties. At both lidar sites, comparable results of the peak value of ash concentration in the upper layer were obtained ($\sim 95 \mu\text{g m}^{-3}$). The obtained values are much lower than those obtained in Germany ($\sim 900 \mu\text{g m}^{-3}$) (Ansmann et al., 2010). Lower ash concentrations and smaller average particle sizes estimated from the Angström exponent may be due to larger distance from the volcano. As some agglomerated ash particles with sizes up to $70 \mu\text{m}$ were found in particle samples collected during airborne measurement, the truncation of the particle size at $10 \mu\text{m}$ AED by the laser particle counter may have introduced a considerable systematic uncertainty in the relationship between the aerosol backscatter coefficient and the TPM content, which may affect the entire concentration profile.

To conclude, results show that lidar-based remote sensing can be used to reliably confirm the presence, altitude and concentration of volcanic ash with lower concentrations and at lower altitudes, where satellite imaging is not feasible. In such cases, lidar-based remote sensing may be of importance in the decision making process regarding the closure of airspace due to the possible presence of volcanic ash plume.

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References

- Ansmann, A., Tesche, M., Groß, S., Freudenthaler, V., Seifert, P., Hiebsch, A., Schmidt, J., Wandinger, U., Mattis, I., Müller, D., and Wiegner, M.: The 16 April 2010 major volcanic ash plume over central Europe: EARLINET lidar and AERONET photometer observations at Leipzig and Munich, Germany, *Geophys. Res. Lett.*, 37, L13810, doi:10.1029/2010GL043809, 2010.
- Casadevall, T. J.: The 1989–1990 eruption of Redoubt Volcano, Alaska: impacts on aircraft operations, *J. Volcanol. Geoth. Res.*, 62, 301–316, 1994.
- Delene, D. J., Rose, W. I., and Grody, N. C.: Remote sensing of volcanic ash clouds using special sensor microwave imager data, *J. Geophys. Res.*, 101(B5), 11579–11588, 1996.
- European Centre for Medium-Range Weather Forecasts, <http://www.ecmwf.int>, access: 31 May 2010.
- Flentje, H., Claude, H., Elste, T., Gilge, S., Köhler, U., Plass-Dülmer, C., Steinbrecht, W., Thomas, W., Werner, A., and Fricke, W.: The Eyjafjallajökull eruption in April 2010 – detection of volcanic plume using in-situ measurements, ozone sondes and lidar-ceilometer profiles, *Atmos. Chem. Phys.*, 10, 10085–10092, doi:10.5194/acp-10-10085-2010, 2010.
- Gao, F., Bergant, K., Filipič, A., Forte, B., Hua, D.-X., Song, X.-Q., Stanič, S., Veberič, D., and Zavrtanik, M.: Observations of the atmospheric boundary layer across the land-sea transition zone using a scanning Mie lidar, *J. Quant. Spectrosc. Ra.*, 112, 182–188, 2011.
- Garratt, J. R.: *The atmospheric boundary layer*, Cambridge University Press, Cambridge, UK, 1992.
- He, T.-Y., Gao, F., Stanič, S., Veberič, D., Bergant, K., Dolžan, A., and Song, X.-Q.: Scanning mobile lidar for aerosol tracking and biological aerosol identification, *Proc. SPIE 7832*, 78320U, doi:10.1117/12.868387, 2010.
- HYbrid Single-Particle Lagrangian Integrated Trajectory: http://www.arl.noaa.gov/HYSPLIT_info.php, access: 16 November 2010.
- Klett, J. D.: Stable analytical inversion solution for processing lidar returns, *Appl. Optics*, 20, 211–220, 1981.
- Larsen, G.: The eruption of the Eyjafjallajökull volcano in 1821–1823, Science Institute Research Report RH-28-99, 13 pp., Reykjavík, 1999.
- Lensky, I. M. and Rosenfeld, D.: Clouds-Aerosols-Precipitation Satellite Analysis Tool (CAPSAT), *Atmos. Chem. Phys.*, 8, 6739–6753, doi:10.5194/acp-8-6739-2008, 2008.
- Meteosat Data: <http://www.eumetsat.int>, access: 8 December 2010.
- Oppenheimer, C., Pyle, D. M., and Barclay, J.: *Volcanic Degassing*, The Geological Society, Bath, UK, 2003.
- Pieri, D., Ma, C., Simpson, J. J., Hufford, G., Grindle, T., and Grove, C.: Analyses of in-situ airborne volcanic ash from the February 2000 eruption of Hekla Volcano, Iceland, *Geophys. Res. Lett.*, 29, 16, doi:10.1029/2001GL013688, 2002.
- Riley, C. M., Rose, W. I., and Bluth, G. J. S.: Quantitative shape measurements of distal volcanic ash, *J. Geophys. Res.*, 108, 2504, doi:10.1029/2001JB000818, 2003.
- Russell, P. B., Bergstrom, R. W., Shinozuka, Y., Clarke, A. D., DeCarlo, P. F., Jimenez, J. L., Livingston, J. M., Redemann, J., Dubovik, O., and Strawa, A.: Absorption Angstrom Exponent in AERONET and related data as an indicator of aerosol composition, *Atmos. Chem. Phys.*, 10, 1155–1169, doi:10.5194/acp-10-1155-2010, 2010.
- Sanderson, K.: Out of the ashes, *Nature*, 544–545, 2010a.
- Sanderson, K.: Questions fly over ash-cloud models, *Nature*, 464, p. 1253, 2010b.
- Sassen, K., Zhu, J., Webley, P., Dean, K., and Cobb, P.: Volcanic ash plume identification using polarization lidar: Augustine eruption, Alaska, *Geophys. Res. Lett.*, 34, L08803, doi:10.1029/2006GL027237, 2007.
- Schumann, U., Weinzierl, B., Reitebuch, O., Schlager, H., Minikin, A., Forster, C., Baumann, R., Sailer, T., Graf, K., Mannstein, H., Voigt, C., Rahm, S., Simmet, R., Scheibe, M., Lichtenstern, M., Stock, P., Rüba, H., Schäuble, D., Tafferner, A., Rautenhaus, M., Gerz, T., Ziereis, H., Krautstrunk, M., Mallaun, C., Gayet, J.-F., Lieke, K., Kandler, K., Ebert, M., Weinbruch, S., Stohl, A., Gasteiger, J., Groß, S., Freudenthaler, V., Wiegner, M., Ansmann, A., Tesche, M., Olafsson, H., and Sturm, K.: Airborne observations of the Eyjafjalla volcano ash cloud over Europe during air space closure in April and May 2010, *Atmos. Chem. Phys.*, 11, 2245–2279, doi:10.5194/acp-11-2245-2011, 2011.
- Sigmundsson, F. and Höskuldsson, Á.: Develop instruments to monitor volcanic ash fallout, *Nature*, 466, p. 28, doi:10.1038/466028b, 2010.
- Taylor, H. E. and Lichte, F. E.: Chemical composition of Mount St. Helens volcanic ash, *Geophys. Res. Lett.*, 7(11), 949–952, 1980.
- US Standard Atmosphere, 1976, US Government Printing Office, Washington, D.C., 1976.
- Wilks, D. S.: *Statistical Methods in the Atmospheric Sciences*, Academic Press, Burlington, MA01803, USA, 1995.
- Witham, C. S.: Volcanic disasters and incidents: A new database, *J. Volcanol. Geoth. Res.*, 148, 191–233, 2005.