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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 detected at ground level using in-situ measurements during the night of 17 April 2010, but was not observed via lidar-based remote sensing due to the presence of clouds at lower altitudes while the streaming height of ash-loaded air masses was above 5 km a.s.l. The second arrival of volcanic ash on 20 April 2010 was detected by both lidar-based remote sensing and airborne in-situ measurement, revealing two or more elevated atmospheric aerosol layers above Slovenia. Identification of samples from ground-based in-situ and airborne in-situ measurements based on energy-dispersive X-ray spectroscopy confirmed that a fraction of particles was volcanic ash from the Eyjafjallajökull eruption. We performed simulations of airflow trajectories to explain the arrival of the air masses containing volcanic ash to Slovenia.

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

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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).

During and after the eruption of Eyjafjallajökull volcano, the presence and characteristics of volcanic ash have been monitored Europe-wide 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). 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 the trajectories of air masses reaching Slovenia.

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2 Synoptic situation over Europe after the eruption

Considering the eruption time of Eyjafjallajökull volcano and the distance between Iceland and Slovenia, the synoptic situation over Europe will be focused on the days from 15 April 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 was slowly moving 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 satellite images, air masses containing volcanic ash reached the air space over Slovenia during the night of 17 April 2010 (Meteosat Data, 2010). Since 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.

Between 17 and 18 April, a shallow low-pressure center formed over Northern Italy and moved towards Slovenia, resulting in precipitation, which during the day slowly moved towards eastern parts of Slovenia. 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 direction towards the north-northeast. Due to the second change in wind direction, air masses containing volcanic ash could arrive in Slovenia again, at lower altitudes, on 20 April 2010.

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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. All the measurements were not 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 the permitted flight route. Measurement sites are shown in Fig. 1. As Slovenia is a small country, the measurements represent the status of the presence of volcanic ash in a rather limited circular zone with an approximate radius of 35 km. The distances between each location are listed in Table 1.

3.1 Ground-based measurements

In Ljubljana, concentrations of solid particles (PM_{10}) and sulphur dioxide (SO_2) in the air as well as the chemical composition of precipitation are monitored routinely by Slovenian Environment Agency. In order to determine whether volcanic ash that was expected to be streaming above Slovenia had been deposited and had potentially any impact on local environment, the collected data has been studied in detail for the period from 10 April to 3 May 2010, starting well before the Eyjafjallajökull eruption and ending after the volcanic ash had spread all over Europe, including Slovenia.

Concentration of PM_{10} particles was continuously monitored at two different heights (ground level and between 1.5 and 4.0 m above the ground) using a tampered element oscillating micro-balance[1]. Monitoring results are shown in Fig. 2 (left). Although the average values throughout the ash monitoring campaign were lower than the daily limit value (50 $\mu g\,m^{-3}$) and were consistent with the average annual concentration, an increment in the data for the period of 16–26 April (about 30 $\mu g\,m^{-3}$) in comparison to the data of 11–15 April (lower than 20 $\mu g\,m^{-3}$) can clearly be seen. In addition, on

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

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certain days (16, 19–22, 24 and 26 April) the maximum hourly concentration of PM_{10} was found to exceed 60 µg m^{-3}. In parallel to the monitoring of particle concentrations, particle sampling was also performed from 10 to 18 April 2010 using sequential particle sampler with PM_{10} and $PM_{2.5}$ reference inlets. Detailed spectral and chemical analysis of the samples is presented in Sect. 4.

Concentrations of SO_2 were continuously monitored using the ultraviolet fluorescent SO_2 analyzer[2], not only in Ljubljana but also in Nova Gorica. Results are shown in Fig. 2 (right). From 11 to 17 April, the average concentration of SO_2 increased linearly from 0.2 µg m^{-3} to 2.1 µg m^{-3}, then decreased linearly until 22 April to 1.0µg m^{-3} (with the exception of a single peak of 1.9 µg m^{-3} on 21 April). The maximum hourly concentration peaked above 8.0 µg m^{-3} on 16, 24 and 26 April. However, all the peak values were below the hourly concentration limit (350 µg m^{-3}) and consistent with the average annual concentration of SO_2.

Monitoring of ash deposition and concentration in the precipitation depended on its occurrence, which was predominant in the periods 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^-, NO_3^- and SO_4^-) in the precipitation samples was checked by ion chromatography using an analytical column[3] 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 spectrometer[4]. Among these, only the presence of F^-, which is usually below the detection limit and is an indication of the presence of aerosols from volcanic activities (Oppenheimer et al., 2003), was observed. The highest hourly concentration of F^- ions was found to be 0.45 mg L^{-1} on 17 April. The highest deposition of F^- ions was 6.64 mg m^{-2} day^{-1} on 18 April 2010.

[2]Model 8850, Teledyne Monitor Labs Inc., USA.

[3]IonPac AS14 analytical column, Dionex Corporation, USA.

[4]ICP-MS, Thermo Electron Corporation, USA.

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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.) (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 recorders[5] in combination with C++ (Linux) based data acquisition software.

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 CET[6]). 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 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). Throughout the lidar operation, meteorological data (temperature, humidity, air pressure and wind velocity) was also monitored by the weather stations co-located to both lidar sites. Meteorological conditions on 20 April were quite similar to that of 19 April as well as to clear sky conditions before 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. Atmospheric extinction coefficient calculated by the Klett method (Klett, 1981) was used to display the atmospheric optical properties. The lidar ratio in the calculation

[5]TR40-160 transient recorder, Licel, Germany.

[6]CET – Central Europe Time, 1 h ahead of Coordinated Universal Time (UTC).

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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).

Representative atmospheric extinction profiles obtained from measurements using the Nova Gorica lidar on 20 April 2010 are shown in Fig. 4, with an offset of 0.05 km^{-1} between successive profiles. Each profile is a result of an observation with a 10 min integration time (about 6000 laser shots). One profile in the morning, two profiles in the afternoon and three profiles in the evening were selected, considering the performing time of the measurements. From the atmospheric extinction profiles, the presence of elevated aerosol layer at an altitude of about 3 km can be clearly seen throughout the entire day. Another elevated aerosol layer at a lower altitude of about 1.5 km can be identified in the evening measurements. To show the variation of these two elevated aerosol layers, the magnitude of the peak atmospheric extinction and its altitude in all of the measured profiles are plotted in Fig. 5. For the upper aerosol layer, the peak value of the atmospheric extinction was found to be smaller in the afternoon (average value of $0.0194 \pm 0.0019\ km^{-1}$) than in the morning (average value of $0.0434 \pm 0.0035\ km^{-1}$) and in the evening (average value of $0.0365 \pm 0.0030\ km^{-1}$), implying that the aerosol concentration was decreasing in the morning and then increasing again in the afternoon. Comparing the variation of the peak values of the atmospheric extinction for the two layers in the morning and evening, the afternoon data manifests much larger changes, which shows that atmospheric conditions were more turbulent in the afternoon and that atmospheric motion was strong during the measurement period. In general, peak altitude of the upper aerosol layer showed a decreasing trend, as it decreased from 3.15 km (average value) in the morning to 2.77 km (average value) in the evening, after atmospheric motion subsided (Fig. 5, right). The lower aerosol layer initially appeared in the atmospheric extinction profile at 16:50 CET. Its corresponding peak value increased from afternoon (average value of $0.0263 \pm 0.0024\ km^{-1}$) to evening (average value of $0.0482 \pm 0.0036\ km^{-1}$). The height of the aerosol layer increased linearly with time from 0.60 km (lowest value in the afternoon) to 1.75 km (highest value in the evening).

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Time series of atmospheric extinction profiles with 1-h intervals obtained from measurements using the Otlica lidar during the night of 20 April 2010 is shown in Fig. 6, with an offset of 0.1 km^{-1} between successive profiles. Each profile is a result of an observation with a 5 min integration time (about 6000 laser shots). All the profiles show the presence of two elevated aerosol layers (at about 2.6 km and 1.7 km). The aerosol density distribution in these two elevated aerosol layers changed with time, which is the result of the interaction of atmospheric structure and atmospheric motion. The variation of the altitude of the atmospheric extinction peaks and of the magnitude of the peak atmospheric extinction are shown in Fig. 7. For the upper aerosol layer, the peak value of the atmospheric extinction 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). However, its average value increased from $0.1386 \pm 0.0037\,km^{-1}$ at around 20:00 CET to $0.2204 \pm 0.0086\,km^{-1}$ at around 01:00+1 CET, implying that the aerosol concentration gradually increased during the night. Its altitude varied between 2.35 km and 2.65 km with an average value of 2.514 km. For the lower aerosol layer, the peak value of the atmospheric extinction increased almost linearly with time from 0.05 km^{-1} to 0.30 km^{-1}. After 23:00 CET, the peak atmospheric extinction value of the lower aerosol layer exceeds the peak value of the upper layer, indicating that at this time the aerosol loading in the lower layer was larger than that in the upper layer. The altitude of the atmospheric extinction peak decreased linearly from 1.78 km at 20:00 CET to 1.60 km at 21:00 CET, then increased again to about 1.95 km at 00:50+1 CET. Average peak altitude of the lower aerosol layer is 1.77 km.

Comparison of atmospheric extinction profiles simultaneously obtained from Nova Gorica and Otlica lidars (i.e. profiles of Fig. 4e and Fig. 6b, taken at 21:40 CET) revealed, in addition to the agreement in the elevation of the atmospheric extinction peaks, a relationship between the peak values of the atmospheric extinction (α). Data shows inverse proportionality to the atmospheric extinction ratio to the ratio of the wavelength (λ) used,

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$$\frac{\alpha_{\text{Otlica}}}{\alpha_{\text{NovaGorica}}} = \frac{\lambda_{\text{NovaGorica}}}{\lambda_{\text{Otlica}}} \simeq 3,$$

which implies that both lidar systems were probing the same layer consisting of the same type and concentration of scatters (Ansmann et al., 1990).

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. 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 used as the input to a laser particle counter[7], which 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 µm. The other half of the conveyed air was manually directed to impinge upon a plastic glue-coated surface (adhesive tape), which was replaced at regular elevation intervals. Three additional adhesive tapes were fixed on the aircraft wing struts to sample the aerosols in the entire air column. The aircraft performed a spiral climb from 0.61 km to 3.05 km at Divača, followed by a leveled flight from Divača to Vrhnika and a spiral descent at Vrhnika to 1.22 km with a constant rate of ascend and descend of 2.5 m s^{-1}. Every 300 m of altitude, a new adhesive tape was exposed for the next 90 m to a direct, unfiltered stream of air. Sampling rate of the laser particle counter was set to 2 s, with the corresponding step in elevation of 5 m at the adopted ascend/descend rate.

Aerosol concentration profiles for six different AED categories measured above Divača and above Vrhnika are shown in Fig. 8, with an offset of 10.0 µg m^{-3} between successive profiles. Above Divača, aerosol concentration for 0.3 µm (a) and 0.5 µm particles (b) manifests similar features, such as "ripples" below 2 km and clearly visible

[7]Lighthouse 3016 IAQ Particle Counter, Lighthouse Worldwide Solutions, USA.

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layers at 1.1 km and 1.5 km. Concentration profiles of 1.0 µm (c) and 2.5 µm particles (d) show a distinct aerosol layer at 2.2 km. Below 2 km, the profile of 2.5 µm particles shows the presence of three additional layers (at 0.6 km, 0.9 km and 1.2 km) with particle concentrations exceeding 1.0 µm in each layer. Concentration profiles of 5.0 µm particles (e) manifests more "ripples" except for a clearly visible peak at 0.85 km with a magnitude of 13.0 $\mu g\,m^{-3}$. The peak at this altitude can also be seen in the distribution of 10.0 µm particles (f) with similar magnitude. Moreover, distribution of 10.0 µm 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 µm (a), 0.5 µm (b) and 1.0 µm particles (c) were much smoother above Vrhnika, showing only a small peak at 2 km. Distribution of 2.5 µm particles (d) has more layers, the most prominent ones being those at 1.9 km and 2.8 km. The concentration for 5.0 µm particles (e) 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 $\mu g\,m^{-3}$) were rather small. However, from the distribution of 10.0 µm particles (f) 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 $\mu g\,m^{-3}$.

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 $40 \sim 50\,\mu g\,m^{-3}$ 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. 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

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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-analysis[8]. 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.

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 sampler 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 performed on 20 April 2010, adhered particles of volcanic ash were found at all altitudes with a peak surface density of approximately 36 particles mm^{-2} (47% of all particles in the sample) at 2.2 km a.s.l. Ash particle candidates were found to be predominantly agglomerated

[8] JEOL 5500 LV, JEOL Ltd., Japan.

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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 adhesion tape samples, 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). Simulation results of 48-h airflow trajectories are shown in Fig. 12 using the ECMWF model (3-h temporal resolution) and in Fig. 13 using the HYSPLIT model (6-h temporal resolution).

Based on the forward trajectories of air masses originating over Iceland (63.63° N, 19.62° W) immediately after the eruption (between 21:00 UTC and 04:00+1 UTC on 15 April 2010) using the ECMWF model (Fig. 12, left) and the HYSPLIT model (Fig. 13, left), air masses carrying volcanic ash were expected to initially reach Slovenia from the north at relatively high altitudes (> 5 km) during the night of 17 April 2010. 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 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.

Backward trajectories at different altitudes were also simulated by both models, where in the case of the ECMWF model the endpoint was only Ljubljana and in the case of HYSPLIT model, multiple endpoints included Ljubljana, Nova Gorica and Divača. Simulations show that, occasionally for a short while trajectories of air-masses

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from Iceland did appear at certain altitudes. However, the phenomenon responsible for the observed arrival of volcanic ash seems to be the continuous streaming of air-masses from the north at altitudes below 6 km for more than 15 h from midnight of 19 April until the afternoon of 20 April 2010, which was present in the simulation results using both models. Examples of the backward airflow trajectories ending at 12:00 UTC on 20 April 2010 are presented by ECMWF in Fig. 12 (right) and by HYSPLIT in Fig. 13 (right). Air masses arrived over Britain, France, Germany, Switzerland, Austria and northern Italy, which are the areas that had already experienced the presence of volcanic ash (Meteosat Data, 2010). 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. After 21 April 2010, there was no direct air-mass trajectories from Iceland that could have carried volcanic ash to Slovenia.

6 Discussions and conclusions

The first arrival of volcanic ash to Slovenia was observed in satellite images on 17 April 2010 (Meteosat Data, 2010). At this time due to local weather conditions and high streaming altitudes, the ash was detected only through the analysis of the precipitation. 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 condition over Europe, especially in the Alpine region. The second arrival of volcanic ash, which could not be predicted from satellite images due to lower ash concentrations and lower streaming altitudes, was observed by lidar-based remote sensing and airborne measurements. The type and density of the aerosol content in the atmosphere was established by spectral and chemical analysis of the collected particle samples.

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All the phenomena observed in ground-based measurements are not yet fully understood, one being that the concentrations of PM_{10} and SO_2 on 23–26 April were even higher than on 17–21 April, when volcanic ash was expected to reach Slovenia. Another unclear issue is that a small amount 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 ash particles (Fig. 3). F^- anions were no longer present in the precipitation on 3 May 2010.

Based on the results of lidar-based remote sensing, we can conclude that the same type of scatters were detected in two elevated aerosol layers by two lidar systems, located about 20 km apart. Differences in the altitudes of the detected layers, especially the lower one, are expected to be due to terrain configuration. Airborne measurement yielded peak values for the TPM content of about $40 \sim 50\,\mu g\,m^{-3}$ 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 is reasonable to establish a relationship between the atmospheric extinction coefficient (α) and the TPM content (c) (Wilks, 1995). Using simultaneous measurements of TPM over Divača and atmospheric extinction over Nova Gorica, the correlation between c and α was found to be linear, $c = (2870 \pm 13)\alpha$, with a correlation coefficient of 0.51 (Fig. 14). In the fit, α was constrained to zero for the limiting case when the TPM is zero. A step of $0.01\,km^{-1}$ in atmospheric extinction for 1064 nm laser thus corresponds to aerosol concentration of about $28.7\,\mu g\,m^{-3}$. 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. 15. Peak value of ash concentration in the upper layer was higher above Otlica ($\sim 115\,\mu g\,m^{-3}$) than above Nova Gorica ($\sim 90\,\mu g\,m^{-3}$), and at both sites much lower than in Germany ($\sim 900\,\mu g\,m^{-3}$) (Ansmann et al., 2010), which may be due

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to larger distance from the volcano. The uncertainties due to the presumed lidar ratio ($R = 55 \pm 5$ sr) and the error of the linear fit in correlation analysis are presented as error bars. We identified two additional sources of uncertainties which are not quantitatively taken into account in Fig. 15. The first is due to truncation of the particle size by the laser particle counter in the airborne measurement at 10 µm AED, while in the simultaneously collected aerosol samples, some agglomerated particles were found with sizes up to 70 µm. The uncertainty may affect the entire concentration profile. The second source of uncertainty is due to the non-coaxial configuration of the lidar systems. Due to their overlap functions (complete overlap zone starts at the range of 1 km at Otlica and 0.5 km at Nova Gorica), the comparison of the absolute values of the lidar return signals below 2 km altitude is not possible.

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 might be of importance in the decision making process regarding the closure of air-space due to the possible presence of volcanic ash plume.

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Table 1. Distance between measurement sites.

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

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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	> 500 m	> 1000 m

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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_2O_3	SiO_2	K_2O	Na_2O	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%	/

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Fig. 1. Map of Slovenia indicating the measuring locations of the campaign. At Nova Gorica and Otlica lidar-based remote sensing was 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 and back (dashed line). The inlayed map of Europe indicates the position of Slovenia and Iceland.

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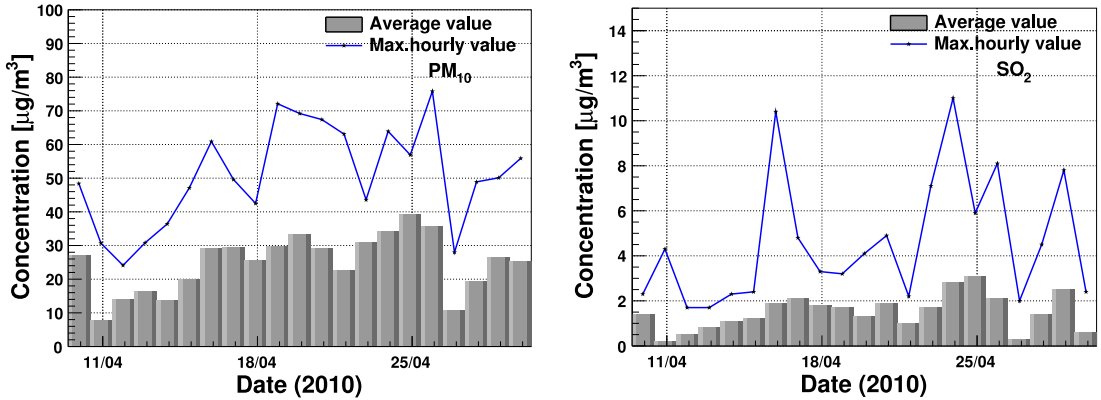

Fig. 2. Monitoring results of the concentration of solid particles (PM_{10}, left) and sulphur dioxide (SO_2, right) in the air in Ljubljana from 10 April to the end of April 2010.

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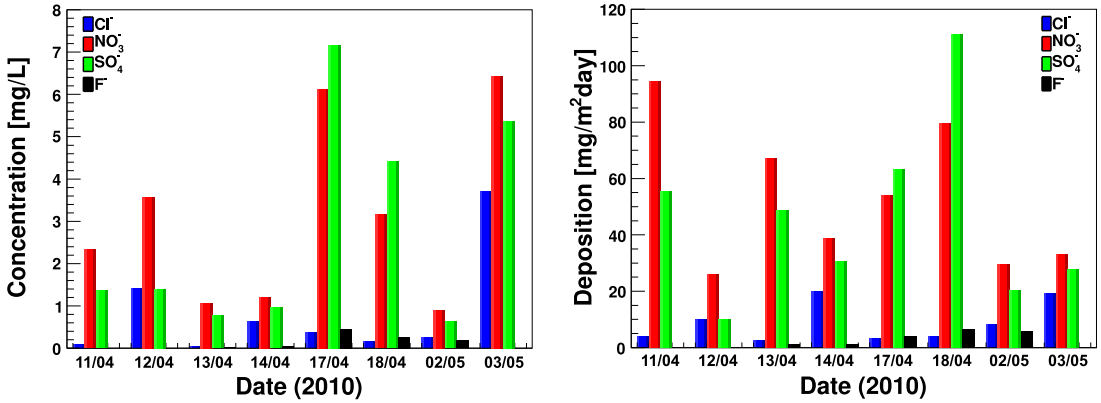

Fig. 3. Results of the composition analysis of precipitation in Ljubljana between 10 April and 3 May 2010. Left: concentration of various anions (Cl^-, NO_3^-, SO_4^- and F^-) in the precipitation; Right: deposition of various anions (Cl^-, NO_3^-, SO_4^- and F^-) in the precipitation.

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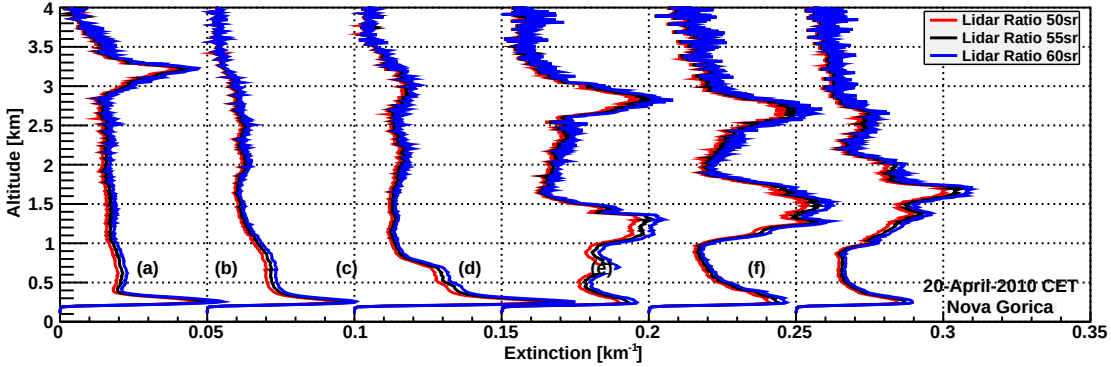

Fig. 4. Examples of atmospheric extinction profiles obtained from measurements by the Nova Gorica lidar on 20 April 2010, with an offset of 0.05 km^{-1} between successive profiles. **(a)** denotes the atmospheric extinction at 10:38, **(b)** at 15:41, **(c)** at 16:33, **(d)** at 20:41, **(e)** at 21:40 and **(f)** at 22:40. All times refer to CET.

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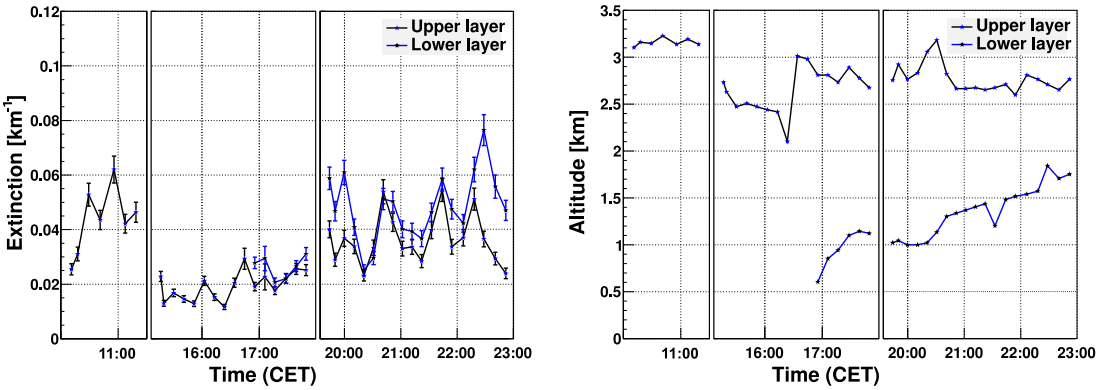

Fig. 5. Variation of peak atmospheric extinction values (left) and variation of the peak altitudes (right) in the profiles measured by the Nova Gorica lidar on 20 April 2010.

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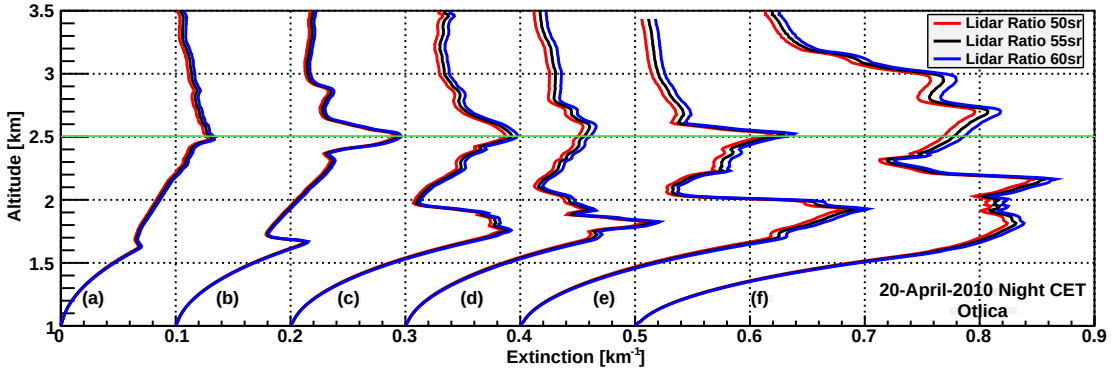

Fig. 6. Time series of atmospheric extinction profiles obtained from measurements by the Otlica lidar during the night of 20 April 2010, with an offset of 0.1 km^{-1} between successive profiles. **(a)** denotes the atmospheric extinction at 20:40, **(b)** at 21:40, **(c)** at 22:40, **(d)** at 23:40, **(e)** at 00:40 and **(f)** at 01:40. All times refer to CET.

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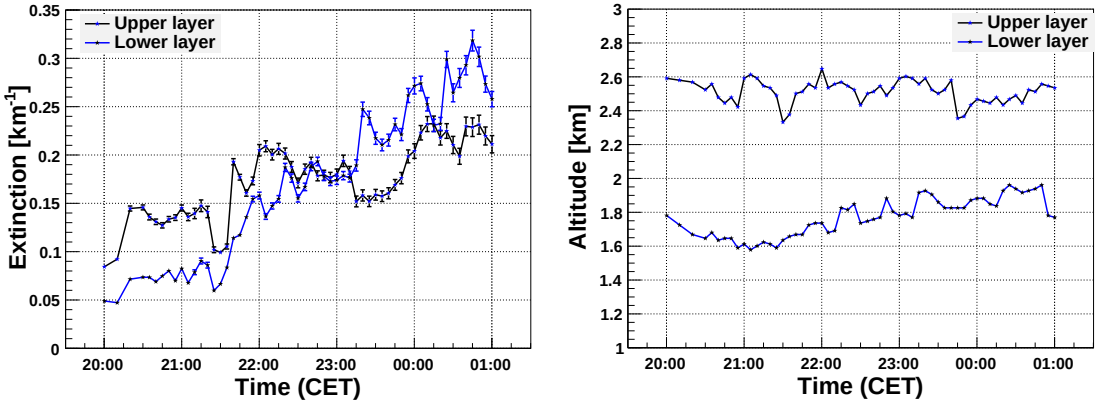

Fig. 7. Variation of peak atmospheric extinction values (left) and variation of the peak altitudes (right) in the profiles measured by the Otlica lidar on 20 April 2010.

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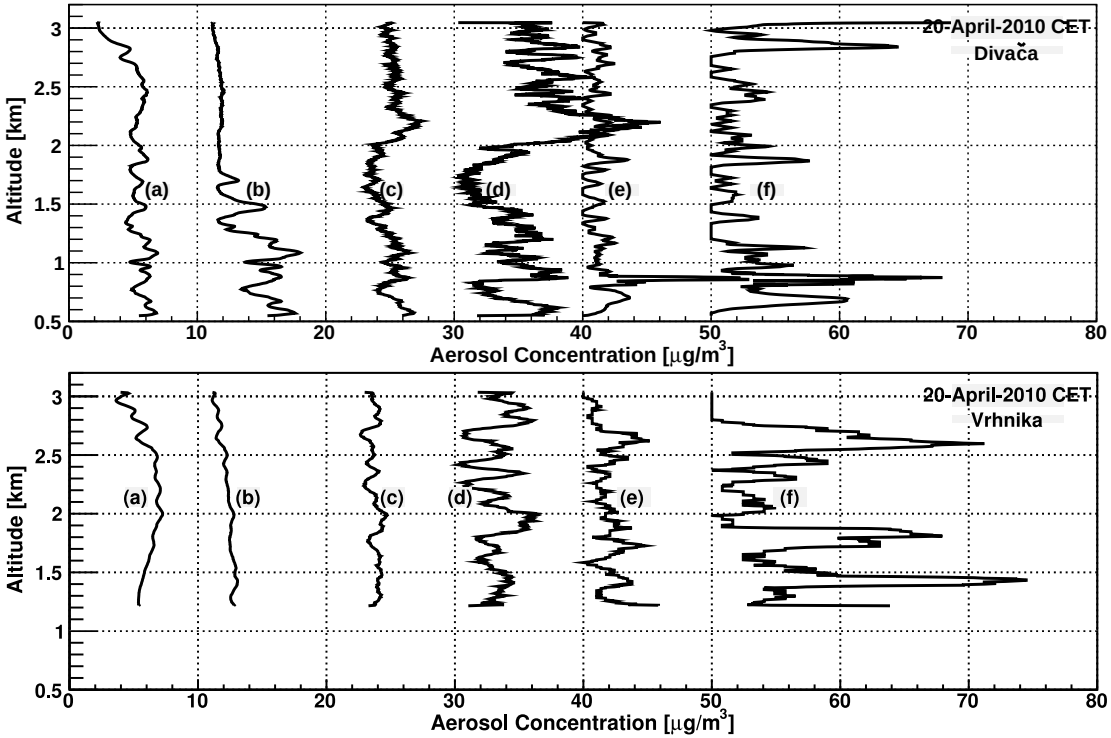

Fig. 8. Aerosol concentration profiles for six different aerosol dynamic equivalent diameters measured on 20 April 2010 above Divača (top) and above Vrhnika (bottom). In both plots, **(a)** denotes the aerosol concentration profiles for 0.3 μm particles; **(b)** for 0.5 μm particles; **(c)** for 1.0 μm particles; **(d)** for 2.5 μm particles; **(e)** for 5.0 μm particles; **(f)** for 10.0 μm particles.

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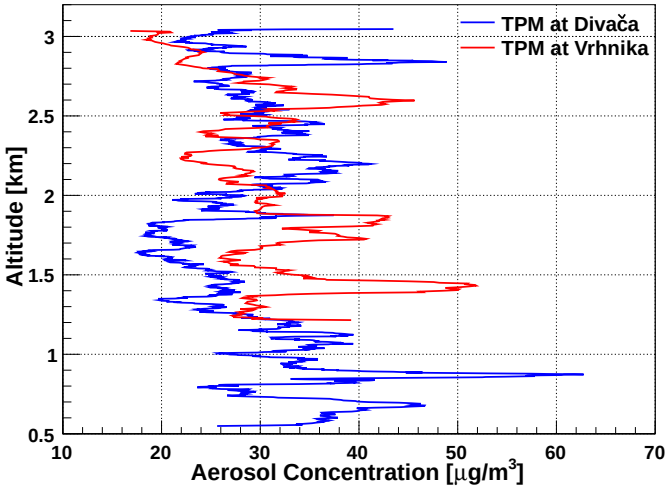

Fig. 9. Profiles of the total particle matter content in the atmosphere above Divača (blue) and above Vrhnika (red) on 20 April 2010 as obtained from the airborne measurements.

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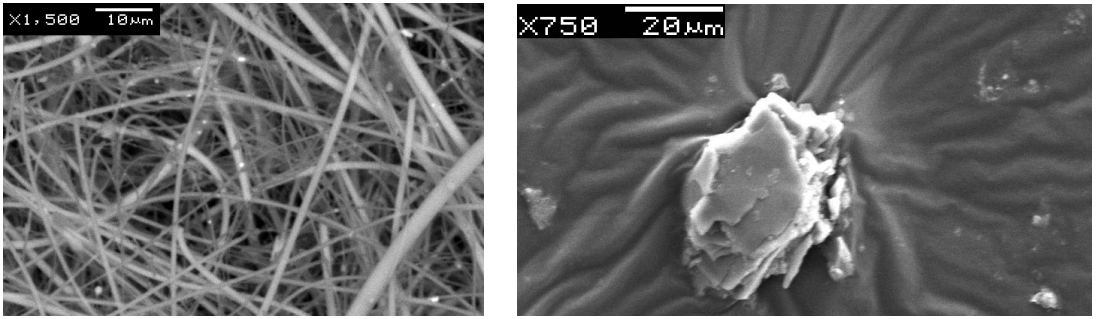

Fig. 10. SEM images of aerosol particles obtained from ground-based and airborne measurements. Left: image of a filter used in the sequential sampler on 17 April 2010 (1500-fold magnification). Bright points on the filter were found mostly to be ash particles. Right: image of an agglomerate ash particle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

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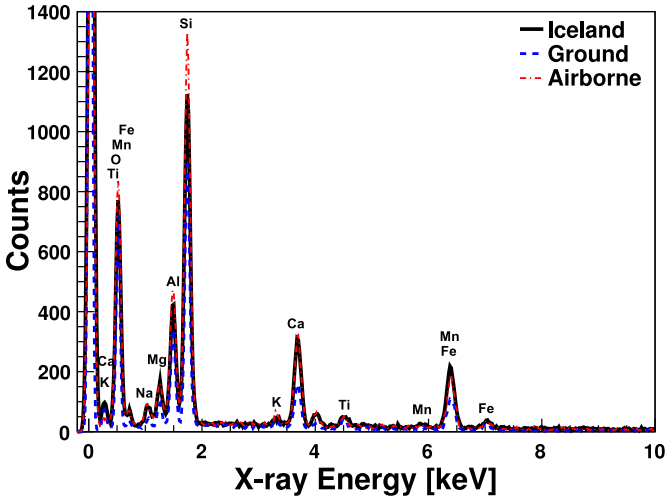

Fig. 11. Energy dispersive X-ray spectrum of the reference ash sample obtained from Iceland (full black line) and the ash sample collected in the ground-based (dashed blue line) and airborne (dotted red line) measurements in Slovenia. The reference sample was collected in the southern part of Iceland,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

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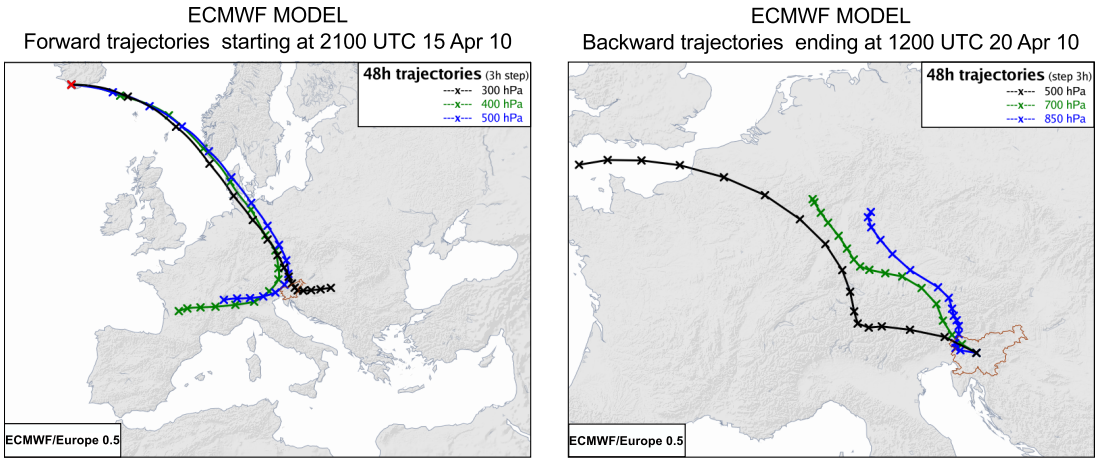

Fig. 12. Air flow trajectories for a 48-h time interval with 3-h temporal resolution obtained from the ECMWF model. Left: forward trajectories for air masses originating near the Eyjafjallajökull volcano starting on 15 April 2010 at 21:00 UTC; Right: backward trajectories of air masses reaching Ljubljana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

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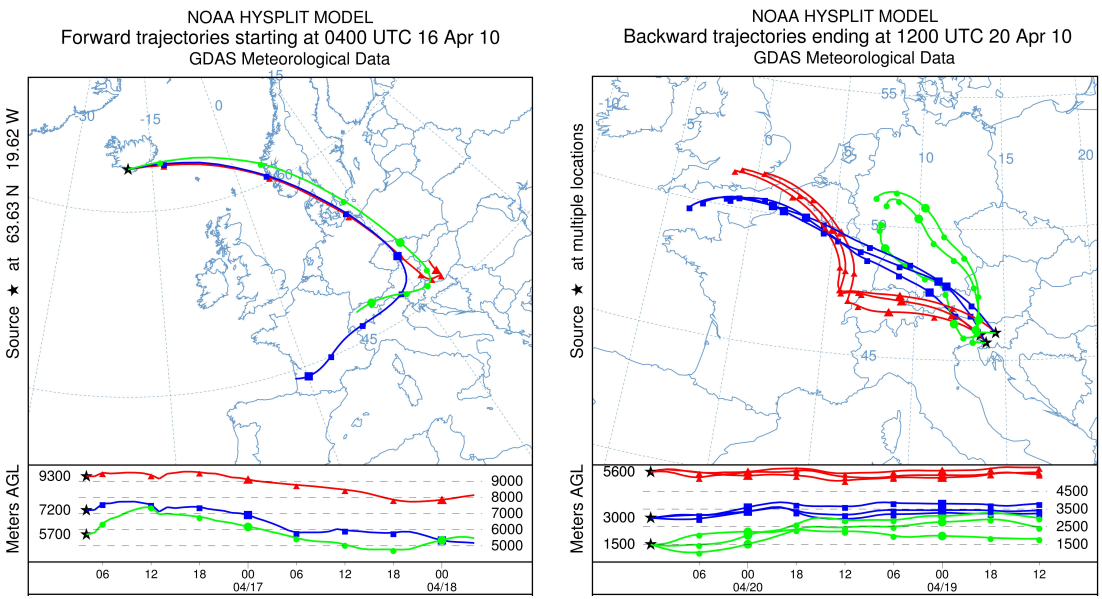

Fig. 13. Air flow trajectories for a 48-h time interval with 6-h temporal resolution obtained from the HYSPLIT model. Left: forward trajectories for air masses originating near the Eyjafjallajökull volcano starting on 16 April 2010 at 04:00 UTC; Right: backward trajectories of air masses reaching Ljubljana,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

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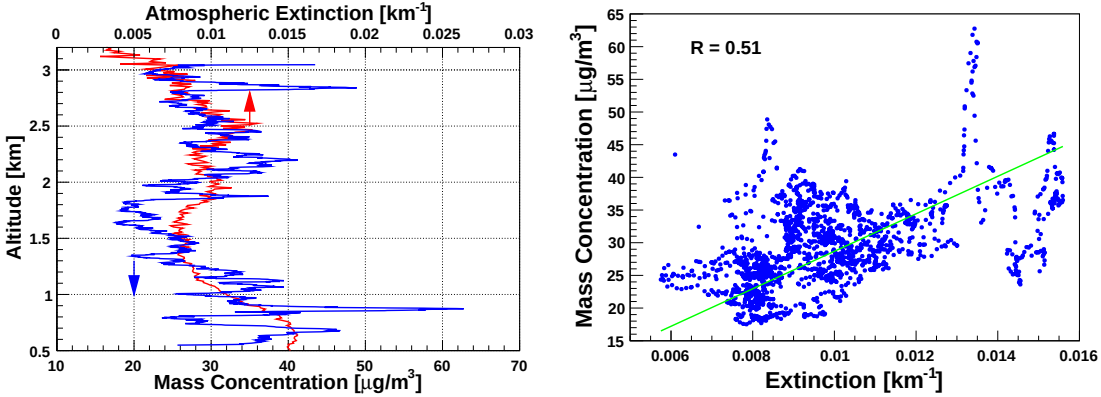

Fig. 14. Total particle matter (TPM) content from the airborne measurement and atmospheric extinction obtained by lidar were found to be correlated. The correlation can be seen qualitatively from the overlap of the TPM (c) profile over Divača and atmospheric extinction (α) profile over Nova Gorica simultaneously measured at 15:20 CET on 20 April 2010 (left). The correlation between c and α was found to be linear, $c = (2870 \pm 13)\alpha$, with a correlation coefficient of 0.51 (right). In the fit, α was constrained to zero for the limit case when the TPM is zero.

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Monitoring presence and streaming patterns of Icelandic volcanic ash

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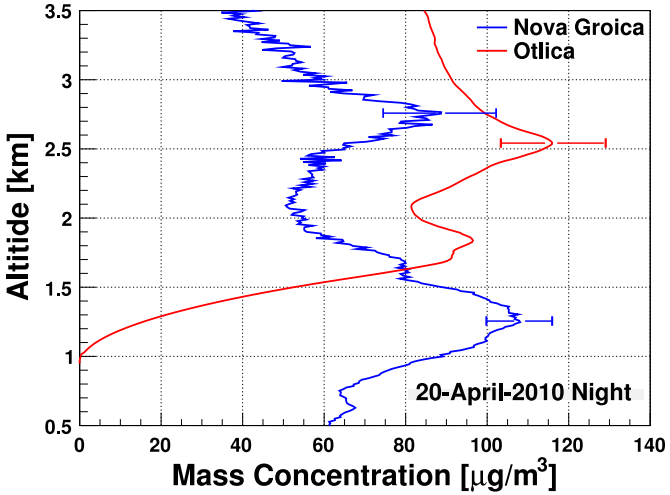

Fig. 15. Mass concentrations of volcanic ash content in the atmosphere above Nova Gorica and Otlica during the first half of the night on 20 April estimated from the approximate 4-h average of the corresponding atmospheric extinction profiles. Error bars denote uncertainties due to the lidar ratio assumption ($R = 55 \pm 5$ sr) and the error of the linear fit in correlation analysis. For the sake of clarity the error bars are plotted at the peak locations in the complete overlap zone only.

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