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Airborne in-situ investigations of the Eyjafjallajökull volcanic ash plume on Iceland and over north-western Germany with light aircrafts and optical particle counters

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ABSTRACT

During the time period of the eruption of the Icelandic volcano Eyjafjallajökull in April/May 2010 the Duesseldorf University of Applied Sciences has performed 14 research flights in situations with and without the volcanic ash plume over Germany. In parallel to the research flights in Germany three measurement flights have been performed by the University of Iceland in May 2010 over the western part of Iceland. During two of these flights the outskirts of the eruption plume were entered directly, delivering most direct measurements within the eruption plume during this eruptive event. For all the measurement flights reported here, light durable piston-motor driven aircrafts were used, which were equipped with optical particle counters for in-situ measurements. Real-time monitoring of the particle concentrations was possible during the flights. As different types of optical particle counters have been used in Iceland and Germany, the optical particle counters have been re-calibrated after the flights to the same standard using gravimetric reference methods and original Eyjafjallajökull volcanic ash samples. In-situ measurement results with high spatial resolution, directly from the eruption plume in Iceland as well as from the dispersed and several days old plume over Germany, are therefore presented here for the first time. They are normalized to the same ash concentration calibration standard. Moreover, airborne particles could be sampled directly out of the eruption plume in Iceland as well as during the flights over Germany.

During the research flights over Iceland from 9 May 2011 to 11 May 2011 the ash emitted from the vent of the volcano turned out to be concentrated in a narrow well-defined plume of about 10 km width at a distance of 45–60 km away from the vent. Outside this plume the airborne ash concentrations could be proved to be below 50 μ g m⁻³ over western Iceland. However, by entering the outskirts of the plume directly the research aircraft could detect ash concentrations of up to 2000 μ g m⁻³.

On the other hand, the ash plume, which was analysed by research flights over Germany several thousand km away from the eruption vent, appeared to be significantly structured in horizontal and vertical directions. Different sub-plumes could be found. Peak concentrations of more than 330 $\mu g\ m^{-3}$ could be detected.

The results of the measurements within the ash plume over Germany were compared with the predictions of the London VAAC model. The range of ash concentrations found by the research aircraft in Germany were not in conflict with the calculations of concentration regimes by the London VAAC model. However, the in-situ measurements performed by the research aircraft were able to deliver information about the structure and composition of the ash plume, which could not be covered by the dispersion model.

Therefore, light piston-motor driven aircrafts equipped with optical particle counters proved to be a very versatile tool for the real-time in-situ determination of the spatial extension of volcanic ash plumes, the ash particle size distributions and the particle mass concentrations. Moreover, all these parameters could be measured with a high horizontal and vertical spatial resolution. Therefore, these

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kinds of measurements can deliver immediate data for the validation and verification of dispersion models and can give direct in-situ information additional to LIDAR measurements and satellite observations. As the piston-motor driven aircrafts are able to operate even at elevated volcanic ash concentrations they can provide valuable ash concentration results for air traffic safety.

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1. Introduction

The Icelandic volcano Eyjafjallajökull erupted on 20 March 2010 in the South of Iceland. The eruption period can be characterized by two different phases: an effusive flank eruption of basalt (20 March to 12 April 2010) and an explosive summit eruption of trachyandesite (14 April to 24 May 2010, Sigmundsson et al., 2010). Large amounts of ash were emitted as melting water from the glacier on the volcano got in contact with the ascending magma. Because of north-westerly winds, ash particles and volcanic gases were transported to western Europe. The plume was detected and tracked by satellite observations (Zehner and Prata, 2010). The Volcanic Ash Advisory Centre (VAAC) of the Meteorological Office London released regularly the results of model calculations for the ash dispersion over Europe as maps of expected ash concentration (Lisk, 2010). Due to strict aviation safety regulations concerning volcanic ash clouds, the airspace over much of Europe was closed from 15 April to 21 April 2010. This caused the cancellation of a huge number of flights and had a severe impact not only on the aviation industry, but also on the economy in general (Perkins, 2010). Later on the airspace was mostly opened again, but local airspace closures took place several times again until mid of May (e.g. 9 May 2010 over Munich, 3 May 2010 over Ireland, 10 May 2010 over Austria).

During this period a large number of different scientific measurement campaigns were performed to get information on the nature and occurrence of the ash plume. Volcanic ash plume dispersion modelling, especially the prediction of the London VAAC, together with the results of the measurement campaigns formed the basis for the decision of the national governments, if the airspace should be closed or not. The measurements comprised for example LIDAR measurements (Pappalardo et al., 2010; Tesche et al., 2010; Groß et al., 2010; Miffre et al., 2010; Flentje et al., 2010), Satellite observations (Stohl et al., 2011), groundbased in-situ measurements (Wurzler et al., 2010; Schäfer et al., 2010; Emeis et al., 2011), as well as balloon (Flentje et al., 2010) and aircraft based measurements (Weber et al., 2010a,b; Schumann et al., 2011; Bukowiecki et al., 2011, Jentink and Velthoven, 2010; Eliasson, 2010; Andrey et al., 2010; Lolli et al., 2010). Aircraft based measurements are of special interest, because they allow of sampling of the ash plume with a high temporal and spatial resolution.

The Duesseldorf University of Applied Sciences (German: Fachhochschule Düsseldorf, FHD) performed aircraft measurements in order to detect the temporal and spatial variation of the plume over north western Germany, and observed size distribution and mass concentration of the ash plume particles. In parallel the University of Iceland performed in-situ aircraft measurements at the Eyjafjallajökull eruption site in Iceland by entering the outskirts of the plume directly and investigated by research flights the degree of potential ash particle pollution in western Iceland, where most of the inhabitants live. The activities of both Universities were performed in parallel operation. However, after the measurement campaigns the instruments of both Universities were calibrated to the same standard and the data were subject to a joint analysis.

2. Measurement equipment

2.1. Aircraft

The airplanes used for the measurement were light aircrafts. A "Flight Design CT" was used in Germany and a "Cessna 206" in Iceland (see Fig. 1). Both aircrafts are equipped with a piston motor with air filter. Piston-motor driven airplanes are safer than jet engine aircrafts to fly in ash plumes, because the air filter of the piston motor can protect the engine from the ash and the piston motor is operating at much lower temperature than a jet engine, thus minimizing the effect of ash melting in the engine. Moreover, piston-motor driven aircrafts have a low minimum cruising speed that allow of measurements with high spatial resolution.

2.2. Optical particle counter

Optical particle counters (OPC) were used for in-situ ash concentration determination with both aircrafts. The measurement principle of these OPC's can be summarized as follows: Ash contaminated air is pumped through the OPC where the particles





Fig. 1. Aircrafts used for the measurements. (a) Flight Design CT: velocity range $90-260 \text{ km h}^{-1}$, measurement velocity $100-120 \text{ km h}^{-1}$, maximum operation distance 1500 km, typical maximum operation altitude 4500 m. (b) Cessna 206: velocity range $110-260 \text{ km h}^{-1}$, measurement velocity $135-160 \text{ km h}^{-1}$, maximum operation distance 1500 km, typical maximum operation altitude 5400 m.





Fig. 2. Eruption plume of the Eyjafjallajökull on 11 May 2010 as seen from the Cessna measurement aircraft: (a) photo taken from north-western position, 10 km distance from the plume and (b) photo taken from western position, 15 km distance from the plume.

cross a continuous laser beam. Every single particle causes a scattering/diffraction of the laser beam. This is recorded by a detector that counts the particles. Moreover, scattering/diffraction intensity of the laser beam is a measure for the size of the particles. From that, the mass can be calculated, provided the density of the particles is known. In this paper we assumed a mean mass density of 2.65 g cm⁻³ for the coarse mode ash particles as it is recommended by EUFAR (2010) for Eyjafjallajökull volcanic ash. More details can be found in (Weber et al., 2006a, 2008; Heim et al., 2008).

The measurements over north western Germany were performed with an OPC Grimm EDM 107 instrument installed at the Flight Design CT aircraft. The laser of the OPC operates at a wavelength of 660 nm. It measures the particle number concentrations in the size range between 250 nm and 32 µm. The particle number concentration range is $0-2*10^6$ particles per litre, that means, even single particles can be counted. The particle loaded air was fed through an isokinetic inlet into the OPC. The good transfer efficiency of the inlet also for relative large particles into the OPC has to be stressed (approx. 50% cut off at about 6 μm particle size, see Section 4, Fig. 21). The isokinetic inlet was designed by the FHD for an air stream velocity of about $90-120 \text{ km h}^{-1}$. The particle concentrations given in this paper are corrected by the particle capture efficiency function (given by the difference of the two functions in Fig. 21). As it could be assumed the ash plume over Germany did not contain particles >15 μm due to the long travelling distances from Iceland (more than 3000 km). So in order to enhance the accuracy of the measurements, the particle sampling over north western Germany was performed for PM10¹, PM2.5 and PM1. The measurements on Iceland were performed with an OPC from Turnkey Instruments Ltd. (Type: DustMate), mounted in the Cessna 206 aircraft. The instrument was connected to the outside-air ventilation intake of the aircraft with a hose where from the OPC sampled air with its inbuilt pump. The laser of this OPC operates at a wavelength of 670 nm. It measures the particle number concentrations in the size range between 300 nm and 20 µm. It can deliver on-line results for TSP-concentrations (Total Suspended Particles) and mass concentrations for PM10, PM2.5 and PM1. During the measurement flights in the Eyjafjallajökull eruption plume on Iceland only the important value for air safety, TSP, was recorded.

The appropriate values for the mean mass density and refractive index to be used for the determination of the mass concentrations of volcanic aerosol are currently matter of discussions (EUFAR, 2010, Bukowiecki et al., 2011; Schumann et al., 2011) Therefore we choose to calibrate the OPC data directly with an independent reference method: Suspended volcanic ash collected in the vicinity of the Eyjafjallajökull was simultaneously measured within laboratory experiments by both OPCs and sampled on filters of gravimetric reference particle measurement systems. The deviation between the gravimetric reference measurement and the Grimm OPCs was only about 11% during this laboratory experiment for PM10 and the present field data were corrected accordingly (see Section 4). Analogous in-field experiments near the Eyjafjallajökull in the post-eruption resuspended ash plume delivered similar calibration results. Details about the re-calibration can be found in the Section 4. All the ash mass concentration data shown here are corrected by this re-calibration. This is of importance for an accurate comparison with the limit values for the aviation, which the aviation authorities agreed on.

2.3. SO₂ passive UV-DOAS measurement system

The UV-DOAS system, which was used, is based on an UV Spectrometer with a spectral range from 280 to 400 nm. Details of the DOAS principle can be found elsewhere (Galle and Oppenheimer, 2002; Platt and Stutz, 2008). The telescope of the UV-DOAS system was mounted outside the fuselage of the aircraft and was directed vertically into the sky. The UV-DOAS system measures the SO_2 -column density above the telescope by determining the integral SO_2 absorption with the scattered skylight as UV source.

2.4. CO₂ measurement system

 ${\rm CO_2}$ concentrations were measured with a non-dispersive infrared (NDIR) gas analyser (Licor LI-840). It is based upon a single path, dual wavelength and infrared detection system. The analyser was connected to the outlet of the OPC, which was equipped with a filter to retain the particles.

2.5. Single particle analysis

The particles collected on the PTFE filter of the OPC outlet were analysed by Computer Controlled Scanning Electron Microscopy (CCSEM). The filters were fixed on a carbon pad for analysis and were coated with a 20 nm carbon layer. Secondary electron (SE) images and chemical analysis of individual particles were obtained

 $^{^{1}}$ PM10 shall mean particulate matter which passes through a size-selective inlet as defined in the reference method for the sampling and measurement of PM10, EN 12341, with a 50% efficiency cut off at 10 μ m aerodynamic diameter.

with a FEI XL Sirion FEG scanning electron microscope (20 kV/4, 4000 magnification) equipped with an EDAX Energy Dispersive X-ray Spectrometer (EDX). On average 500 particles per filter were analysed. Both morphological and chemical data were treated with the EDAX GENESIS software. Chemical compositions were calculated from measured net intensities after correction for matrix, absorption and fluorescence effects (ZAF corrections). The compositions in tables and diagrams are given either in oxide or atomic weight %.

3. Measurements, results and discussion

3.1. Measurement flights on Iceland 9-11 May 2010

The Icelandic airport operator ISAVIA asked the Engineering Research Institute (ERI) of the University of Iceland to come up with a plan to measure the airspace contamination caused by Eyjafjallajökull. In two weeks ERI had equipped a commercial piston engine Cessna 206 aircraft with a DustMate OPC dustmeter and performed the initial test flights (Eliasson, 2010). The flight route was tracked by GPS. Fig. 2 shows photos of the eruption plume as could be observed from the Cessna aircraft on 11 May 2010.

The three measurement flights were run along very much the same route: From Reykjavik, over Keflavik across the sea parallel to the southern coast in direction to the Eyjafjallajökull and back to Reykjavik (see Figs. 3 and 4, yellow line). They showed very similar results. The plume had a limited width and extension during these days, because of the meteorological conditions (about 10 km width in about 50 km distance from the vent, (compare Figs. 3 and 4)).



Fig. 3. (a) Track of measurement flight on 10 May 2010: Concentration measurements (total suspended particles), dark orange mark on flight track: high ash concentrations (up to 1800 $\mu g \ m^{-3}$) (b) MODIS Satellite photo of 10 May 2010 (NASA-MODIS, 2010).

Concentration outside the well-defined plume was found below 50 $\mu g \ m^{-3}$ everywhere over western Iceland. Plume height was between 3000 and 4300 m variable, as it could be detected visually from the aircraft. The weather was clear sky with occasional drifting water vapour clouds but no rain, wind from NNW 15–20 m s⁻¹ in 3000 m. Insignificant ash pollution (below 50 $\mu g \ m^{-3}$) was observed outside the visible ash cloud.

The results of the three flights can be summarized as follows:

3.1.1. 9 May 2010, measurement flight outside the plume over western Iceland

The flight was performed over western Iceland (similar as in Fig. 3), but keeping always at least 50 km distance from the well-defined visible plume The concentrations over western Iceland found outside the plume were below 50 μ g m⁻³.

3.1.2. 10 May 2010, measurement flight with short penetration into the visible plume

The track of this measurement flight is plotted in Fig. 3a. The measurements showed concentrations outside the plume of below 50 μg m⁻³, same as the day before. This result proved, that the transport of the ash emitted by the volcano was concentrated in a narrow plume throughout these days, which can also be clearly seen in the MODIS picture, Fig. 3b. When the research aircraft

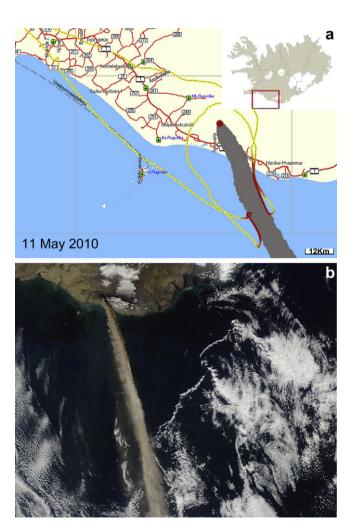


Fig. 4. (a) Track of measurement flight on 11 May 2010: Concentration measurements (total suspended particles), dark orange mark on flight track: high ash concentrations (up to 2000 μ g m⁻³) (b) MODIS Satellite photo of 11 May 2010 (NASA-MODIS, 2010).

entered the western outskirts of the plume on 10 May 2010 in a distance of 15 km away from the vent (see Fig. 3a), the measured concentrations increased dramatically up to values of about 1000 up to 1800 $\mu g m^{-3}$.

3.1.3. 11 May 2010, measurement flight with penetration into the visible plume boundary

On this day the outskirts of the visible plume at a distance of about 45-60 km away from the vent were entered by the research aircraft (see Fig. 4a) for several minutes. Visible means here, that the plume could be detected by the eye without instrumentation from any side and direction. The plume appeared to be inhomogeneous and showed some kind of ash cloud puffs (500 m-2 km diameter) at the boundary, which can be detected at the MODIS picture in Fig. 4b as well. The puffs were entered directly by the aircraft. These measurements appear to be the most direct in-situ measurements reported until now in the eruption plume of the Eyjafjallajökull. The ash concentration was measured, flying through the drifting ash cloud puffs, on the visible cloud boundary. The concentration results varied from 500 μg m⁻³ up to about 2000 μg m⁻³ inside the puffs. These measured concentrations are validated later on in re-calibration experiments with gravimetric reference samplers (see Section 4). The width of the visible plume (horizontal extension) could be determined from the flight track as about 10 km at the time and position of the flight (45-60 km away from the vent). During the flight in the ash cloud puffs the emissions of the eruption could be smelled inside the cabin of the airplane (sulphuric compounds). The impression of the visibility inside the puffs was like flying inside a cloud of black smoke of fire. Colour of the ash cloud was dark grey close to the crater, white on top over the sea, darker below. Air turbulence in cloud was low. Flying in and out of the puffs the aircraft detected no turbulence like the one often is found when aircrafts fly in and out of rainclouds. Aircrafts will not have any immediate problems flying just inside the plume in such cases, and there will be no problems if they keep safe distance from the visible plume cloud.

It should be stressed at this point, that this piston-motor driven aircraft encountered no problems when flying in these high ash

concentrations. The air filter of the airplane engine was inspected afterwards and was not blocked by the ash particles. Some of the ash particles on the air filter were analysed later on with CCSEM. The analysis revealed the presence of bigger particles of more than 100 μm diameter in the ash plume near (15–60 km) to the vent of the Eyjafjallajökull (Fig. 5).

The puffs can be detected on the satellite photos (Figs. 3 and 4) and they seem to survive 200-300 km downwind. These puffs must cause an enormous scatter in concentration values making remote observations of the concentration very difficult. The measurements indicate that $1000-2000~\mu g~m^{-3}$ is the visibility limit in clear weather daylight conditions. Clouds with lower concentration are not readily detectible by the eye without instruments, except perhaps if they are spread out over several kilometres in the in-sight direction. (see Section 3.2). It is therefore interesting to note that the lower limit concentration (red/gray limit) for aircraft used in the later stages of the Eyjafjallajökull crisis was $2000~\mu g~m^{-3}$ since 19 May 2010~(EU, 2010).

3.2. Measurement flights over north western Germany

In the time period from 23 April to 21 May 2010 fourteen measurement flights have been performed by the Duesseldorf University of Applied Sciences.

Four additional flights served for visual observation, photographic recording, referencing to ash free atmosphere as well as transfer of the aircraft.

The flights covered time periods, for which the London VAAC model predicted ash plumes over major parts of Germany, but also periods where the simulations showed no or only local presence of volcanic ash over Germany. The measurement flights gave the opportunity to get spatial highly resolved in-situ particle concentration data. Moreover the particle concentrations could be monitored on-line during the flights. The flight tracks were planned according to the London VAAC model ash predictions, which were released every 6 h.

In the following paragraphs flight trajectories and corresponding measurement results during two different representative

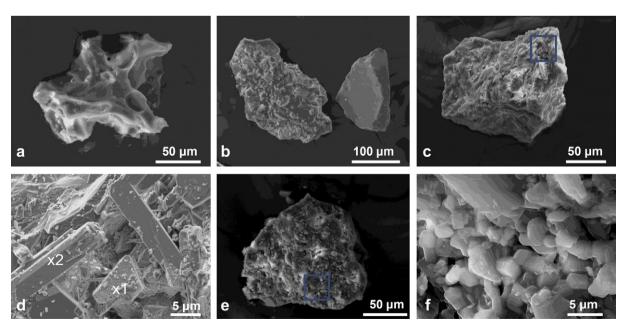


Fig. 5. Structures of particles found on the air filter of the Cessna 206 airplane during the flight in the ash plume on 11 May 2010 showing different morphologies after the flight through the ash plume. The particles were mounted on a carbon pad for analyses: (a) Porous glass particle; (b) left: Crystals in a glassy matrix, right: Crystal fragment (MgFe-silicate: Olivine); (c) Compact particle with crystal rich zones (White square); (d) Crystal rich zone of particle of image c with FeTi-Oxide (x1) and Pyroxenes (x2). Both crystal phases could also be detected by XRD on bulk ash samples from Iceland; (e) Particle dominated by crystals; (f) Detail image of particle (e) showing a crystalline texture.

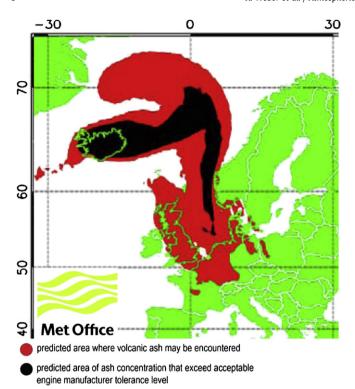


Fig. 6. VAAC prediction for volcanic ash plume over Europe; Red: Predicted area where volcanic ash may be encountered with peak concentrations $> 200~\mu g~m^{-3}$; Black: predicted area of ash concentrations that exceed acceptable manufacturer tolerance levels. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article).

examples of ash plume coverage (with and without ash plume) over Germany are presented. All measurement flights started from the small airfield "Schwarze Heide" (N 51°36′59″, E 6°51′55″) in Germany north of Duesseldorf.

3.2.1. Measurement flight on 18 May 2010 within the "red warning zone" of ash predicted over Germany

On the 18 May 2010 the VAAC simulation showed Germany almost entirely located within a "red warning zone" (see Fig. 6), i.e. a zone, where volcanic ash could be encountered with possible peak concentrations $\geq 200~\mu g~m^{-3}$. In the early morning it looked as if some German airports had to be closed again, because of an assumed coming "black zone" over northern Germany. Therefore, the German Weather Service stipulated measurement flights over the affected regions. FHD prepared two measurement flights with two identical aircrafts and identical OPCs. The ash plume forecast at 06:00 UTC, however, maintained a "red warning zone" over Germany. The measurements of one of the aircrafts showed only very low ash concentration at the position of the flight track. Therefore only the results of the other research aircraft are reported here.

This aircraft was equipped with an OPC EDM 107, an UV-DOAS system and a CO_2 measurement system. The OPC outlet was equipped with a filter to collect the particles taken in during the flight for later analysis.

The aircraft started from the airfield "Schwarze Heide" in the northern part of the Rhein—Ruhr area, headed along the Dutch border in direction of the North Sea, continued towards Hamburg (13:50 UTC) and returned back via Münster to the airfield "Schwarze Heide". VAAC predicted a "red warning zone" for the major part of Germany for the entire flight time (Fig. 6).

Elevated PM10 concentrations were recorded several times during the flight: along the Dutch border between Borken and Twist (see Figs. 7 and 9), and during the following flight track at Leer (11:15), near Bremerhafen (13:10), near Bassum (14:15) and between Glandorf and Altendorf (14:40–15:00) (Fig. 8). These measurement results agreed with backward trajectory calculation with the NOAA HYSPLIT model. These model calculations show clearly (Fig. 10), that the air masses present during the flight time along Dutch border were originating in Iceland. The same day, a measurement aircraft of the Netherlands (Jentink and Velthoven,

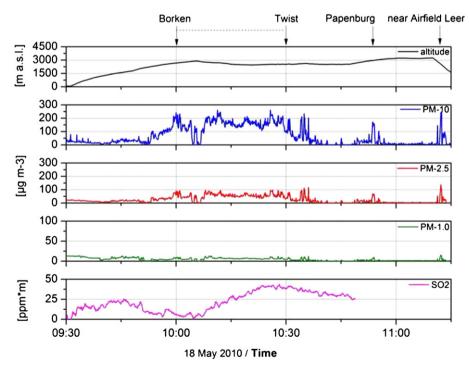


Fig. 7. PM and SO₂-concentrations measured along the Dutch border on 18 May 2011.

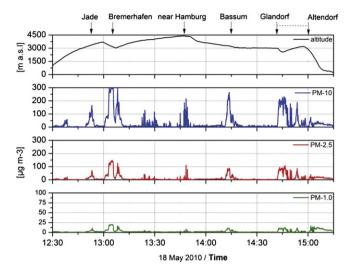


Fig. 8. PM concentrations measured during the flight over northern Germany on 18 May 2011.

2010) recorded also elevated concentrations along the German–Dutch border.

Along the Dutch border PM10, PM2.5, and PM1 showed increased concentrations up to 260 $\mu g \ m^{-3}$ between Borken and Twist (between 10:00 and 10:40 UTC, Fig. 7). For the time periods 10:40-10:50 UTC and 10:55-11:10 UTC the measured concentrations dropped to very low values of a few ug m^{-3} PM10. Evidently during these time periods the aircraft did not encounter parts of the ash plume and measured only particle concentrations of a few $\mu g m^{-3}$, which are typical for the normal atmosphere above the planetary boundary layer (PBL). The difference between PM10 and PM2.5 concentrations in the flight period along the Dutch border between 10:00–10:40 UTC indicate that particles with diameters larger than 2.5 µm had to be present (PM10). The OPC was able to deliver a complete analysis of the particle size distribution every 6 s. Four typical examples of particle size distributions are shown in Fig. 11 sampled at different times (two size distributions obtained within the ash plume along the Dutch border, two size distributions obtained outside the ash plume). The size distribution obtained from the OPC confirmed the presence of a number of larger ash particles in the plume, more than outside the plume. Moreover it is interesting to see that the size distributions within the plume at the Dutch border are very similar.

A remarkable fact is that the high ash plume concentrations during this flight were encountered already at altitudes slightly above 2500 m (s. Fig. 7). The concentration between the top of the PBL (ca. 1500 m) and the cloud was also significantly higher than expected in the free troposphere (about 09:45 UTC). This might suggest a downward mixing of parts of the ash plume, which was already present at 17 May, towards the PBL. This interpretation is supported by observations of measurements from LIDAR (Groß et al., 2010) and groundbased station networks (Wurzler et al., 2010; Schäfer et al., 2010).

During this flight the particles measured by the OPC were collected on the PTFE filter at the outlet of the OPC. About 500 particles were analyzed by CCSEM.

The chemical compositions of the particles (Table 1) were compared with analyses made from eruption products sampled in the vicinity of the volcano. The analyses CFG_01, 03 and 04 are very close to the bulk ash compositions given by the Nordic Volcanological Center (http://www.earthice.hi.is/page/IES-EY-CEMCOM). CFG_04, 05 and 06 are enriched in silica and depleted in the iron oxide, but all other oxide components are again close to the bulk composition of the ash. The last two particles have similar compositions. The atomic formula obtained after normalization of these analyses to 6 oxygens corresponds to augitic clinopyroxenes, which together with magnetite are present as phenocrysts in the eruption products (confirmed by X-ray diffraction analyses). The majority of the analyses cluster around the average bulk composition of the eruption. This is also confirmed by the ternary diagram of airborne samples taken during the measurement flight along the Dutch border (Fig. 12).

Some examples of images of the airborne ash particles taken during the flight of the research aircraft are shown in Fig. 13. They have different morphology and sizes. Particles coincoidal fracture patterns typical for glass as well as particles with well-developed crystal faces. The SEM images revealed also larger particles with sizes of up to 6.5 μ m confirming the OPC results.

Together with the particle concentration, the gases SO₂ and CO₂, which are typical components of volcanic gas emissions (Oppenheimer et al., 2003; Schmincke, 1998; Weber et al., 2006b), have been monitored during the flight. The SO₂-concentrations (Fig. 7) along the Dutch border were enhanced and showed a certain correlation with PM10. A strong correlation between the measured particle concentration and the measured SO₂

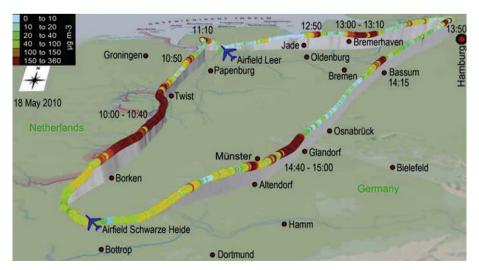
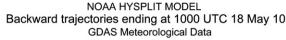


Fig. 9. Track of measurement flight on 18 May 2010 with measured concentration of PM10, colours indicate the concentration range.



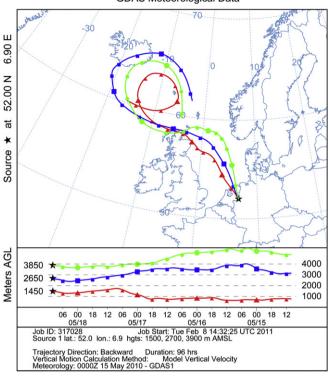


Fig. 10. Backward trajectories from NOAA HYSPLIT model for the German—Dutch boarder on 18 May 2010.

concentration is not to be expected due to the different sampling volumes of the OPC and the UV-DOAS measurements. Whereas the particle concentration is measured in the air stream around the airplane, the UV-DOAS determines the SO₂-column density above the aircraft. Moreover, because of the several thousand km distance from the emitting volcano the gaseous SO₂ compound might undergo different atmospheric dispersion processes compared with the ash particles. These dispersion effects might change with time and location. Bukowiecki et al. found also changing correlations between SO₂ and ash particle concentrations during groundbased measurements at the Jungfraujoch in Switzerland (Bukowiecki et al., 2011). In some cases the ash plume and

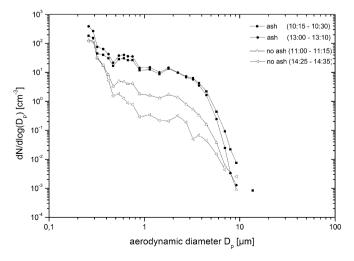


Fig. 11. Particle size distribution of the flight along the German–Dutch boarder and in northern Germany on 18 May 2010.

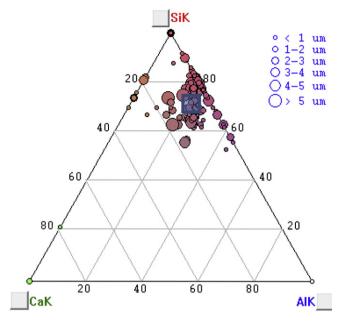


Fig. 12. Ternary diagram of airborne particle samples taken during measurement flight. Circles represent the chemical composition of the airborne particles. The square represents the composition of volcanic ash sampled in the vicinity of the Eyjafjallajökull.

SO₂-plume can separate significantly shortly after the eruption, as it was reported by Kerminen et al. for the 2011 Grimsvötn eruption. However, during the Eyjafjallajökull eruption the high airborne SO₂-concentrations found at the Dutch border by the research aircraft are a clear sign for the presence of air masses that came from the Eyjafjallajökull, as there were no major industrial SO2 sources present in the area of measurements. In photographic pictures, taken near Nordhorn about 10:30 UTC in a westerly direction (Fig. 14), the ash cloud appears as a small brown colored layer at the horizon. It was visible only in horizontal direction, not in vertical direction. Near Papenburg (10:50 UTC) the ash plume had a double layer structure, looking from the aircraft into western direction. Double or multiple layer structures have been detected during the flight by the OPC and visually as well (Weber et al., 2010a). The CO₂ concentrations measured during the flight did not show significant variation (only 3%) due to the plume. This is certainly due to the fact, that CO2 is a major constituent of the standard atmosphere and the measurements have been taken about 3000 km away from the Eyjafjallajökull.

Along he further flight track beyond the Dutch border more ash prannes structures were encountered (Fig. 8). Near Bremerhafen (13:10 UTC) peak concentrations of more than 330 $\mu g \ m^{-3}$ PM10 were observed. The vertical depth of the ash plume determined by spiraling up and down near Bremerhafen was about 300 m, again with a two layer substructure. Whereas the flight track along the Dutch border was over 80 km continuously within an ash cloud, the following encounters were of much shorter duration. This may be due to a patchy structure of ash plumes or because the flight trajectory (rather W–E compared to N–S along the Dutch border) was perpendicular to the length extension of the clouds.

The main result of the flight with the research aircraft was that on the 18 May 2010 ash plumes were present in the airspace over northern Germany at altitudes between 2500 and 4500 m. The small cruising speed of the aircraft (90 km h^{-1} –120 km h^{-1}) allowed to resolve the fine structure of the ash plume, something which is difficult if not impossible for fast flying jets.

Table 1Chemical composition in weight percent of representative samples taken during the flight on 18 May 2011 of research aircraft based on EDX data (here numbered as CFG 0x and Di 0x).

Samples	CFG_01	CFG_02	CFG_03	CFG_04	CFG_05	CFG_06	CFG_07	Ref	Di_01	Di_02
SiO ₂	65.29	59.01	57.06	56.16	50.38	66.27	67.86	57.98	47.33	48.65
Al_2O_3	15.46	12.81	15.69	10.29	13.52	15.92	16.59	14.87	0.81	0.74
Fe_2O_3	6.37	12.43	11.96	12.74	15.73	5.73	5.83	9.75	26.95	28.08
MnO	0.27	0.70	0.43	0.32	0.65	0.00	0.00	0.24	2.73	1.27
MgO	0.00	4.18	1.33	4.48	0.58	1.12	0.92	2.3	11.38	12.41
CaO	2.03	2.80	2.49	9.19	1.73	1.44	1.22	5.5	8.68	6.95
Na ₂ O	5.92	5.22	7.59	5.18	6.30	7.15	4.48	5.01	0.42	1.39
K ₂ O	2.79	1.65	1.35	0.61	1.67	1.96	3.10	1.79	0.00	0.00
TiO ₂	0.98	1.19	1.47	0.74	8.52	0.00	0.00	1.8	0.68	0.51
Total	99.11	99.99	99.37	99.71	99.08	99.59	100	99.37	98.98	100.0

The research aircraft had no problem at all with the encountered ash. No signs of damage were found neither in the engine or on the aircraft's windows, wings or fuselage. A piston-motor driven aircraft is to be expected to have much less problems with ash particles than a jet engine. This is due to several technical factors: the air filter, which is used for piston-motors, is filtering the majority of ash particles out of the air before it enters the motor. Moreover, the parts in a piston motor are moving more slowly as parts in the jet engines and the temperatures reached by the moving parts are considerably lower.

The piston-motor driven aircraft (Cessna 206) performed measurement flights on Iceland beyond the visible boundary of the eruption plume of the Eyjafjallajökull in Iceland in May 2010. The aircraft successfully resisted concentration levels about 2000 $\mu g \ m^{-3}$ (Eliasson, 2010).

3.2.2. Measurement flight on 19 May 2010 with no plume predicted over north-western Germany

For the late afternoon hours of 19 May 2010 there was no ash plume to be expected over north-western Germany according to the VAAC prediction. The only "red warning zone" for central Europe was located over France (Fig. 15). A measurement flight was performed at this day in order to investigate, if remains of the ash plume of the day before were still around. The flight started at the airfield "Schwarze Heide", went to Ochtrup (15:25 UTC) climbing

up above the PBL (see Fig. 16), but no ash particle concentrations were encountered above the PBL between 15:00 and 15:30 UTC (Fig. 17). During the return flight back to the airfield "Schwarze Heide" the research aircraft flew at low altitudes within the PBL. Therefore the particle concentrations typical for the PBL could be measured around 25–100 $\mu g\ m^{-3}$. These measurement results were in agreement with the NOAA HYSPLIT backward trajectory calculations indicating that the air masses during the flight time and flight position did not originate from Iceland (Fig. 18). Therefore on 19 May 2010 the meteorological conditions were favourable to completely clear the troposphere.

3.3. Discussion

The measurement results of the research aircrafts reported here gave valuable informations about the spread and concentration range of the Eyjafjallajökull ash plume by using OPC instruments. The aircraft measurements on Iceland could give ash concentration data over western Iceland and directly in the outskirts of the plume. Moreover from these measurements it could be concluded, how refined to a comparative small width the ash plume was over Iceland during the period of 9–11 May 2011.

On the other hand, the aircraft measurements over Germany could give a detailed and spatial highly resolved mapping of the plume several thousand kilometers away from the vent. These

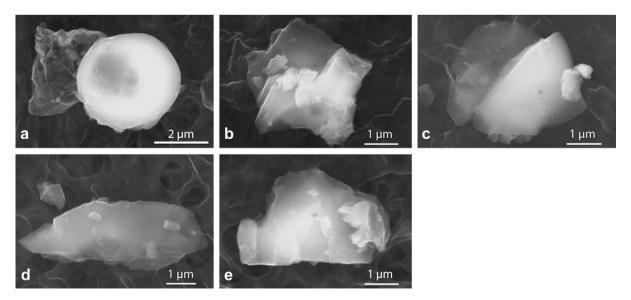


Fig. 13. SEM images from airborne particles taken out of the plume during the measurement flight over northern Germany on 18 May 2010 (flight track, see Fig. 9): (1) Silicious spherule (3.7 μm) with a chemical composition close to the bulk composition of ash from Eyjafjallajökull; (b) Silicious glass particle (3.9 μm) with a chemical composition similar to bulk composition of ash from Eyjafjallajökull; (c) Silicious glass particle (3.3 μm) with a chemical composition of ash from Eyjafjallajökull; (d) two silicious particle (6.5 and 1.2 μm) with a chemical composition close to the bulk composition of ash from Eyjafjallajökull, but without Ti and Mg; (e) Silicious particle (4.5 μm) with a chemical composition close to the bulk composition of ash from Eyjafjallajökull, but without Fe and Ti and less Mg.

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Fig. 14. Ash plume visible during the measurement flight in horizontal direction on 18 May 2010.

aircraft measurements revealed a more structured plume as it could be seen from the dispersion model of the VAAC. As the in-situ OPC data described in this paper were re-calibrated to the same gravimetrical reference ash standard, these kinds of aircraft measurements seem to be appropriate for validation and verification of dispersion models. Moreover the OPCs used for these aircraft measurements can take advantage from the fact, that they are fast and have a very low detection limit: They can count even single particles. Another advantage of these aircraft measurements is that they can deliver real-time in-situ results. These results are not dependent from eruption source parameters or dispersion conditions, as it is the case for dispersion models.

Moreover, aircraft measurements can successfully supplement results from satellite observations: Whereas satellite observations reveal broader plume features under good visibility conditions and cannot give a real vertical resolution of the plume, the aircraft

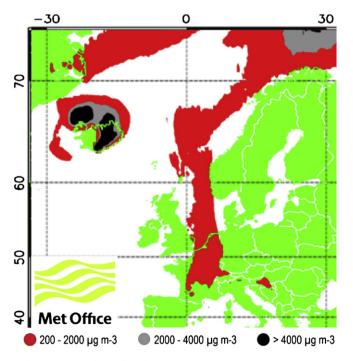


Fig. 15. VAAC prediction for volcanic ash plume over Europe on 19 May 2010.



Fig. 16. Track of measurement flight on 19 May 2010 with measured concentration of PM10, colours indicate the concentration range.

measurements are able to detect plume concentration variations even in vertical extent by spiraling flight patterns.

LIDAR measurements during the Eyjafjallajökull eruption period showed also very strong temporal and spatial variations of the ash plume over Europe (see e.g. Pappalardo et al., 2010) as it could be detected by the aircraft measurements in this paper. It is interesting to compare the data of the aircraft measurements of this paper with results of other aircraft measurements in this time period. Bukowiecki et al. (2011) reported an ash layer at an altitude of 3500 m a.s.l. over the Swiss plateau, which was detected during the flight of the DIMO aircraft on 18 May 2010, equipped with OPCs as well. The average mass concentration was reported here as $320 \, \mu g \, m^{-3}$, which is in the same order as the measurement results reported in this paper for the peak concentrations in northern Germany. Schumann et al. (2011) reported a measurement flight as well over Germany on the 18 May 2011 in the morning hours. These authors found about 110 $\mu g\ m^{-3}$ ash concentration over Hamburg and up to 340 $\mu g m^{-3}$ in southern Germany. Therefore, concerning the slightly different flight times and flight paths, the concentrations reported by the different research aircrafts can be regarded as very consistent. Research aircraft measurements seem to be a very suitable tool, to validate general features of dispersion models and to deliver additional detailed information about the local structure and density of the ash plume additional to the more general dispersion model calculations. This proved to be true also during

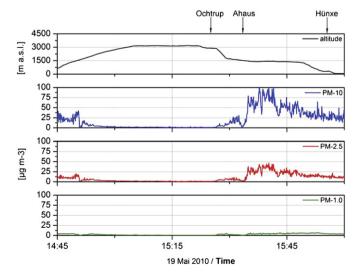


Fig. 17. PM concentrations on 19 May 2010 at the Rhein—Ruhr area between Airfield Schwarze Heide and Ochtrup.

NOAA HYSPLIT MODEL Backward trajectories ending at 1500 UTC 19 May 10 GDAS Meteorological Data

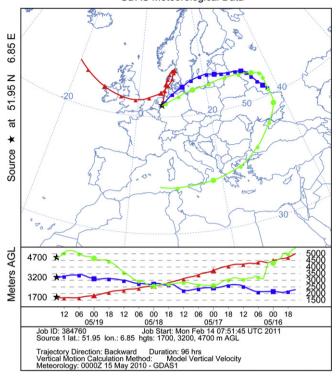


Fig. 18. Backward trajectories from NOAA HYSPLIT model for the German-Dutch boarder on 19 May 2010.

the Grimsvötn eruption 2011, where the assessment of low ash concentrations over western Iceland by a research aircraft contributed to a re-opening of the Keflavik airport despite of higher ash concentration predictions by the dispersion model for a larger area (Eliasson et al., 2011).

4. Calibration and quality assurance aspects

The appropriate values for the mean ash particle mass density and refractive index to be used for the determination of the mass concentrations of volcanic aerosol are currently matter of discussions. A value of around 2.65 g cm⁻³ for the density of volcanic Eyjafjallajökull ash is proposed by the European Facility for Airborne Research (EUFAR, 2010) and used for the airborne measurements of this study as a first order for on-line results. In order to achieve maximum accuracy we calibrated the OPCs in post-flight experiments independently with gravimetric reference samplers using resuspended Eyjafjallajökull ash. The gravimetric reference method is prescribed in European legislation for ambient air particle measurements (European Parliament, 2008). In this method the particle polluted air is pumped through filters and causes a particle load on the filter, which is determined by weighing of the filters before and after the particle loading.

We used a linear wind tunnel for these calibration experiments equipped with an additional particle suspension device, which was able to feed the ash particles, which were collected in the vicinity of Eyjafjallajökull, at a constant rate into the wind tunnel. This horizontal wind tunnel (25 m long) belongs to the Institute for the Research on Hazardous Substances, Bochum, Germany and was operated at a slow wind speed of 0.6 m s⁻¹ especially for this gravimetric comparison. Details of the wind tunnel are explained in Dahmann et al. (2004). Constant steady states for different ash concentrations could be realized within the wind tunnel, depending on the ash suspension rate and wind speed. The ash concentrations were simultaneously measured by both OPCs and sampled on the filters of the gravimetric reference particle measurement systems. In this way the different OPCs could be normalized to the same reference standard and a function for the re-calibration of the OPCs and the in-flight measurement results could be achieved. The re-calibration functions regarding PM10 and PM2.5 for the Grimm EDM 107 are plotted in Fig. 19 (a) and (b). The deviation between the gravimetric reference measurement and the Grimm OPC for PM10 was only about 9% during this wind tunnel experiment. The aircraft based field data were corrected accordingly. Analogous in-field experiments near the Eyjafjallajökull in the post-eruption resuspended ash plume and in different ash sedimentation chambers at FHD delivered comparable calibration results. All the ash mass concentration data shown throughout this study were corrected by this re-calibration. This is of importance for the accurate comparison of the measured values with the limit values of aviation safety. Moreover, this re-calibration gives now the opportunity for a better surveillance of the aviation safety limits in a future eruption case.

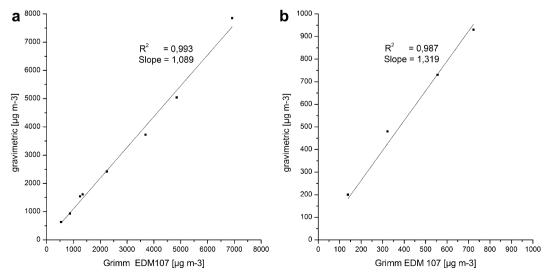


Fig. 19. Correction factors for the OPC Grimm EDM 107 for PM10 and PM2.5 measurements respectively (a) 1.09 and (b) 1.32.

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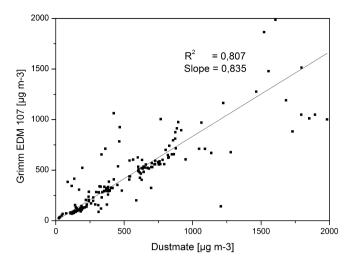


Fig. 20. Correlation between OPC GRIMM EDM 107 and OPC Turnkey Dustmate.

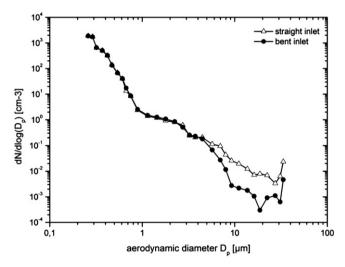


Fig. 21. Difference in sampling efficiencies for particles taken with a straight isokinetic sampling inlet and a 90° bent isokinetic sampling inlet.

The linear wind tunnel experiments were also used as well to establish the correlation between the different OPCs Grimm EDM 107 and Turnkey DustMate. As it can be seen in Fig. 20 the difference between these OPCs turned out to be about 16% only for PM10 within these experiments up to a concentration of 2000 $\mu g\ m^{-3}.$ With the use of the gravimetric reference systems it was therefore possible to normalize the Turnkey DustMate and the Grimm EDM 107 to the same standard.

Fig. 21 shows the evaluation of the collection efficiency of a bent isokinetic sampling inlet in comparison with a straight isokinetic sampling inlet with an optimized sampling efficiency. The 50% sampling efficiency cut off was at about 6 μm particle size for the bent isokinetic sampling inlet. The collection efficiencies in Fig. 21 were determined in wind tunnel experiments at a velocity of about 100 km h^{-1} , corresponding to the measurement velocity of the aircrafts and in a similar way with driving cars in a heavily polluted atmosphere at the same driving speed.

5. Conclusions

Numerous research flights have been performed by the Duesseldorf University of Applied Sciences and the University of Iceland in April and May 2010 in the Eyjafjallajökull ash plume. The University of Iceland covered western Iceland with measurement flights and was able to perform in-situ particle measurements within the ash plume very close the eruption vent of the Eyjafjallajökull. The Duesseldorf University of Applied Sciences performed research flights over north-western Germany during time periods with and without predicted ash plume over Germany. The optical particle counters of both institutions were re-calibrated with Eyjafjallajökull ash to the same gravimetric standard. The measurement flights of the University of Iceland delivered peak concentrations of up to 2000 $\mu g \ m^{-3}$ in the outskirts of the eruption plume $45{-}60 \ km$ away from the vent in southern direction, whereas the particle load outside the plume over western Iceland was less than $50 \ \mu g \ m^{-3}$ throughout the measurement periods.

The research flights of the Duesseldorf University of Applied Sciences more than 3000 km away from the eruption source over north-western Germany, showed an inhomogeneous structure of the ash plume on the kilometer scale, as it can be seen in Fig. 9. Layers of one hundred meters to a few hundred meters vertical extend of ash plume could be identified. Sub-plumes with a horizontal extent of several kilometers to several tenths of kilometers could be found at altitudes between 2500 m and 4500 m. Peak concentrations of more than 330 $\mu g \ m^{-3}$ could be observed. The measured particle concentrations and the observed plume extensions have been compared with the simulation of the Volcanic Ash Advisory Centre (VAAC). At the resolution of the VAAC model, the predictions were not in conflict with the airborne measurement results in following sense: In the situation of 18 May 2010, when the VAAC model predicted a "red warning zone" over Germany, the research aircraft encountered indeed ash concentrations with peak levels higher than 330 μ g m⁻³. On 19 May 2010, when no ash plume was predicted over Germany by the VAAC, indeed the research aircraft did not find ash during the control flight.

However, the research flights revealed a much more structured ash plume over Germany than the VAAC dispersion could predict.

The studies showed that light piston-motor driven aircrafts equipped with OPCs are a very versatile tool for the investigation of the volcanic ash plume. They can easily be equipped with additional measurement systems for e.g. SO₂ measurements und particle sampling for a post-flight analysis in the laboratory. Moreover the piston-motor driven aircrafts seem to be robust and durable enough to fly at even elevated concentrations of about 2000 $\mu g m^{-3}$, which is important for the surveillance of air safety limits. Therefore for the investigation of the eruption plume of a volcano it should be possible to perform orthogonal cross-wind and downwind measurements in the plume in this concentration range, giving an estimate of parameters for plume geometry, particle distribution, particle concentrations and mass flux. Moreover, this study demonstrates that light slow flying aircrafts allow mapping of plumes with high spatial resolution. Therefore they seem to be a very versatile tool for the investigation of volcanic plumes.

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Appendix. Supplementary material

Supplementary material related to this article can be found at doi:10.1016/j.atmosenv.2011.10.030.

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