In vitro evaluation of pulmonary deposition of airborne volcanic ash

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Highlights
- Re-suspension of ash can substantially increase airborne particle concentrations.
- Up to 9% of the ash particle surface area was deposited into the alveolar region.
- Most of the particle surface area was deposited to the head airways.

Abstract
There has been an increasing interest in the effects of volcanic eruption on the environment, climate, and health following two recent volcanic eruptions in Iceland. Although health issues are mainly focused on subjects living close to the eruption due to the high concentration of airborne ash and gases in close vicinity to the volcanoes, the ash may also reach high altitude and get distributed thousands of kilometers away from the volcano. Ash particles used in the studies were collected at the Eyjafjallajökull and Grímsvötn eruption sites. The composition, size, density and morphology of the particles were analyzed and the effect of particle properties on the re-dispersion and lung deposition were studied. The aero-dynamic size and morphology of the particles were consistent with field measurement results obtained during the eruptions. Due to their size and structure, the ash particles can be re-suspended and transported into the lungs. The total surface area of submicron ash particles deposited into the alveolar and tracheobronchial regions of the lungs were 3e9% and 1e2%, respectively. Although the main fraction of the surface area is deposited in the head airways region, a significant amount of particles can deposit into the alveolar and tracheobronchial regions. The results indicate that a substantial increase in the concentration of respirable airborne ash particles and associated health hazard can take place if the deposited ash particles are re-suspended under dry, windy conditions or by outdoor human activity.

1. Introduction
The Eyjafjallajökull volcano eruption took place from April 14 to late May 2010 ejecting around 480 Tg of volcanic ash whereof 80% was airborne tephra, i.e. around 380 Tg (Gislason et al., 2011b; Gudmundsson et al., 2012; Schumann et al., 2011). A second volcano, Grímsvötn, erupted a year later in May 2011. Most of the mass from Grímsvötn was ejected over the first 24 h of the eruption. Both eruptions caused a large release of particles to the environment. The eruption of Eyjafjallajökull and Grímsvötn eruption sites. The composition, size, density and morphology of the particles were analyzed and the effect of particle properties on the re-dispersion and lung deposition were studied. The aero-dynamic size and morphology of the particles were consistent with field measurement results obtained during the eruptions. Due to their size and structure, the ash particles can be re-suspended and transported into the lungs. The total surface area of submicron ash particles deposited into the alveolar and tracheobronchial regions of the lungs were 3e9% and 1e2%, respectively. Although the main fraction of the surface area is deposited in the head airways region, a significant amount of particles can deposit into the alveolar and tracheobronchial regions. The results indicate that a substantial increase in the concentration of respirable airborne ash particles and associated health hazard can take place if the deposited ash particles are re-suspended under dry, windy conditions or by outdoor human activity.
less attention was paid to the environmental pollution and potential health effects of these particles. However, increased PM$_{10}$ (mass of particles smaller than 10 $\mu$m) values were observed in a number of locations around Europe (Bukowiecki et al., 2011; Colette et al., 2011; Donovan and Oppenheimer, 2011; Kerminen et al., 2011; Schäfer et al., 2011). For example, high PM$_{10}$ concentrations up to 140 $\mu$g m$^{-3}$ were measured at Schauinsland and 100 $\mu$g m$^{-3}$ in Freiburg in Germany (Schäfer et al., 2011), and hourly PM$_{10}$ concentrations exceeded 100 $\mu$g m$^{-3}$ in the southern parts of Scandinavia for several hours (Tesche et al., 2012). However, much higher mean daily average PM$_{10}$ value were observed in Reykjavik on June 4, 2010 was about 500 $\mu$g m$^{-3}$ after Eyjafjallajökull eruption (Bodvarsdottir and Sigurdardottir, 2010; Thorsteinsson et al., 2012). After the Grímsvötn eruption the maximum value was measured to be around 200 $\mu$g m$^{-3}$ on May 22, 2011. These values clearly exceed the PM$_{10}$ threshold of 50 $\mu$g m$^{-3}$ set by the European Union air quality directive 2008/50/EC (Schäfer et al., 2011).

The high emissions can have important local and regional implications that affect vegetation, livestock and people especially within Iceland (Graham et al., 1985; Gudmundsson et al., 2008). Health effects of airborne particles depend on the dose, which in case of particles in air is normally derived from the measured particle concentration, the breathing flow rate and the breathing frequency of the individual (Asbach et al., 2009). Airborne fine particles with the aerodynamic diameter below 2.5 $\mu$m and especially ultrafine particles (diameter $<$ 100 nm), with small mass per particle but large surface area per unit mass, have been associated with adverse health effects such as cardiac and respiratory diseases (Dockery, 2009; Katsouyanni et al., 1997; Löndahl et al., 2007; Pope and Dockery, 2006). In particular the health hazard due to prolonged exposure to airborne respirable silica, a major constituent of volcanic ash, is well known (Edmonds et al., 1977; Graham et al., 1985). A population based survey of early health effects of the Eyjafjallajökull eruption was undertaken in Iceland at the end of the eruption (Carlsen et al., 2012). The short-term ash exposure was associated with upper airway irritation symptoms and exacerbation of pre-existing asthma but did not contribute to serious health problems. The exposure did not impair respiratory function compared to controls. Outdoor use of protective glasses and face masks was considered protective against irritation to the eyes and the upper airways.

In this paper we compare the aerosolization of bulk ash particles collected 60 km from the Eyjafjallajökull eruption on 15 April 2010 and 80 km from Grímsvötn eruption on May 22nd 2011. The emphasis of this study is on the characterization, evaluation and comparison of the particulate matter emitted from these two volcanoes. The ash samples collected from the ground during the eruptions consist of small particles adhering to the surface of larger particles. This structure resembles that of typical pharmaceutical dry powder formulation for inhalation, consisting of micronized drug particles with coarse carrier particles (Prime et al., 1997). Therefore, the re-dispersion of the particles and aerosol properties were studied. In addition, in vitro pulmonary deposition of the ash particles from Eyja and Grím were assessed based on their aerodynamic diameter and total number concentration. Furthermore, the surface area of particles corresponding to the deposition in the tracheobronchial and alveolar regions of human lungs was determined with the nanoparticle surface area monitor. This gives valuable information of the health effects of the particles, since the reactivity of the particles within the lung is nowadays known to be related to the surface area and number of particles more than mass (Horwell and Baxter, 2006). The data obtained in these studies were compared to the environmental particle concentrations measured during the eruptions.

2. Materials and methods

2.1. Materials

Sample of the Eyjafjallajökull ash was collected on the main road in south-eastern Iceland, about 60 km east of the summit crater on 15 April 2010 12:30:00 GMT about 30 h after the onset of the summit eruption of the Eyjafjallajökull (Gislason et al., 2011a, 2011b), see Fig. 1. The collection site was located at the southern

![Fig. 1. A map of south Iceland showing the locations of Eyjafjallajökull and Grímsvötn and sites where the settled ash samples were collected (indicated Eyja and Grím).](image-url)
2.2. Aerosolization of the ash particles

Fig. 2 shows the experimental set-up used for the aerosolization of the ash particles. It can be divided into the (1) particle dispersion, (2) dilution and mixing, and (3) sampling sections. The ash particles were dispersed using a fluidized bed aerosol generator (FBAG, Model 3400A, TSI Inc., USA). The system is able to disperse dry powders, with aerodynamic diameter of the particles ranging from 0.15 to 0.4 cm s<sup>−1</sup> corresponding to average air velocity of 0.15 and 0.4 cm s<sup>−1</sup>, respectively. No size classification, i.e. cyclone, was used at the FBAG outlet. The aerosol exiting the FBAG was diluted with dry air at ambient temperature, using a porous tube diluter, to obtain a suitable concentration range for the on-line aerosol analyzers (Lyyränen et al., 2004). To ensure isokinetic sampling the total flow rate was kept at 62.5 L min<sup>−1</sup> during the experiments by adjusting the dilution flow rate down from 47.5 L min<sup>−1</sup> to 32.5 L min<sup>−1</sup> as the aerosol flow rate increased from 15 to 30 L min<sup>−1</sup>.

2.3. Instrumentation and characterization

The determination of the grain size distributions of the samples as collected on the ground was attempted with dynamic light scattering (Zetasizer nanoZS, Malvern Instruments). The water suspensions of particles were sonicated for 60 min before the measurement. Bulk density of the settled ash samples was obtained directly from the ratio of mass and volume, i.e. 20.3 g Eyjafjallajökull and 17.4 g Grímsvötn ash were measured in 25 ml graduated cylinder. Tap density was determined using an ERWEKA tap density analyzer (SVM, Apparatebau GMBH, Heusenstamm, Germany). The bulk and the tap densities were used in the calculation of the Carr’s compressibility index (Staniforth and Aulton, 2007).

The aerodynamic number size distribution and the related geometric mean diameter of the aerosolized ash particles were measured on-line with electrical low-pressure impactor (ELPI, Dekati Ltd., Finland) using isokinetic sampling. The aerodynamic particle size is defined as the diameter of a spherical particle with a density of 1000 kg m<sup>−3</sup>, i.e. it is equivalent to a water droplet having the same aerodynamic properties as the particle (Hinds, 1999). The total particle number concentration in the gas phase was determined with a condensation particle counter (CPC 3776, TSI Inc., USA).

The surface area of the nanoparticles corresponding to the deposition in the tracheobronchial (TB, trap voltage 100 V) and alveolar (AL, trap voltage 200 V) regions of the lungs were determined with a nanoparticle surface area monitor (NSAM, model 3550, TSI Inc., USA) (Shin et al., 2007). A cyclone specifically designed for NSAM was used to remove all particles above 1000 nm, which is the upper limit of the size range that can still be reliably measured with this equipment (Asbach et al., 2009). Particle deposition onto TB, AL and head airways regions (HA) were also calculated from the particle size distributions determined with ELPI using the ICRP respiratory deposition models (Asbach et al., 2009; Hinds, 1999; ICRP, 1994; Leskinen et al., 2012). In the particle lung deposition calculations from ELPI data, mass and surface area size distributions were obtained from the number size distributions using one mode log-normal distribution fitting with the particle density of 2 g cm<sup>−3</sup>. Results were scaled to a PM<sub>10</sub> concentration of 1 mg m<sup>−3</sup> to enable comparison of the results. Furthermore, the hygroscopic growth of the particles can significantly alter particle deposition in lung (Löndahl et al., 2007). However, according to a recent study (Lathem et al., 2011) the hygroscopic growth of volcanic ash particles is very weak as the settled ash edge of the plume which was heading nearly straight east. The samples were collected using a Plexiglas spatula and heavy walled polyethylene bags (Gislason et al., 2011a). The samples were kept in the bags in a dry place and samples intended for experimental and microscopic studies were kept in dessicators. The size of the samples, several kilos, acted in itself as desiccators.

The Grímsvötn samples were collected about 80 km from the crater on 22 May 2011, the 2nd day of eruption, when the density of the airborne ash was at the highest in the town of Kirkjubæjarölklaustur (Fig. 1). The sample was collected from the ground in the town of Kirkjubæjarölklaustur sampled in the dark, within the plume that was heading south–west (Fig. 1). The sample was placed into a heavy wallet plastic bag and shipped immediately for analysis.

Both Eyja and Grím collection sites were selected because these were the closest farm communities (Kirkjubæjarölklaustur town/Grímsvötn, and Álftaver farm community/Eyjafjallajökull) most severely affected by the ash, during the eruptions. Both samples were collected within 30 h of the onset of the eruption before they were rained on. The distance from the eruption was 60–80 km in both cases.
particles typically consist of water insoluble or only very slightly soluble compounds (e.g., silica, iron oxides). The hygroscopicity parameter reported for Eyjafjallajökull ash particles was less than 0.01 corresponding to growth factor value of ca. 1.12 in lungs. Thus, it has been ignored in the calculations.

The morphology of the collected particles was imaged with a field emission low-voltage electron microscope (FE-SEM, Leo Gemini DSM 982) operated at a 2 kV acceleration voltage. The elemental composition of the particles was analyzed using a transmission electron microscope (TEM, Jeol JEM-2100F) with energy dispersion spectrometer (EDS, Thermo Scientific NS7 with Si(Li) crystal) operated at a 200 kV acceleration voltage. The aerosolized samples for the electron microscope analyses were collected directly from the aerosol flow on a holey carbon copper grid (Agar Scientific Inc., S147-4400 Holey Carbon Film 400 Mesh Cu) using an aspiration-based electron microscope sampler (Lyyränen et al., 2009). The dry powder samples of bulk ash particles were prepared on stubs using double sided carbon tabs (SPI Supplies). The particle composition and crystallinity were determined with an X-ray diffractometer (XRD, Bruker D8 DISCOVER). The specific surface area of the samples was determined with nitrogen adsorption isotherms (i.e. BET method) measured with Tristar II 3020 porosity analyzer (Micrometrics Instrument Corp., USA).

3. Results and discussion

3.1. Properties of settled ash particles

Based on the dynamic light scattering analysis both samples were very polydispersive. The Eyja sample consisted of submicron particles with size between 100 nm and 200 nm, while large particles sedimented on the bottom of suspension. The particle size in the Gríms sample could not be measured as it was more polydisperse than Eyja sample. However, the obtained polydispersivity of the samples corresponds well with the SEM images of the volcanic ash particles shown in Fig. 3. The size of the particles sedimented out of the ash plume in close vicinity of Eyja ranged from submicron up to 50 μm while the size of the Grím particles ranged from submicron to 100 μm. In addition, the Grím particles were more faceted than the Eyja particles. The tapped density value and the Carr's compressibility index for Eyja were 1292 kg m⁻³ and 23%, respectively. The corresponding values for Grím were 1183 kg m⁻³ and 25%. Furthermore, the specific surface area (BET) of the settled ash particles was 3.09 m² g⁻¹ for Eyja and 0.53 m² g⁻¹ for Grím. These kinds of values are typical for relatively large and non-porous particles with poor flowability (Staniforth and Aulton, 2007). The difference in the particle properties originates from the differences in the conditions during the particle formation, i.e. the force of explosive vs. effusive activity of the subglacial volcanoes, the distance from the eruption site and the chemical composition of the magma (Donovan and Oppenheimer, 2011). However, in both cases the coarse ash particles were covered with smaller particles adhered to their surface (Fig. 3). A substantial fraction of these fine particles in the ash can cause a significant concentration of respirable airborne particles if the deposited ash is re-suspended under dry, windy conditions or by outdoor human activity (Horwell et al., 2010; Thorsteinsson et al., 2012). An example of such events is the dust storms that took place close to Eyja and affected the air quality as far as 100 km away in the capital Reykjavik (Donovan and Oppenheimer, 2011).

Fig. 3. SEM images of the particles collected from the ground near Eyjafjallajökull (A scale 50 μm, B scale 10 μm) and Grímsvötn (C scale 50 μm, D scale 10 μm).
The X-ray diffraction patterns of the settled ash samples are shown in Fig. 4. There is elemental variation between the Eyja and Grím samples that arises from different magma compositions. The volcanic ash particles from Eyja have substantial silica content and the peaks between 26 and 27° 20 can be assigned to quartz. This is consistent with the composition obtained from the airborne particles in Europe during the eruption (Bukowiecki et al., 2011; Lettino et al., 2012; Petäjä et al., 2012; Schumann et al., 2011). The presence of silicates and iron oxides were also confirmed with TEM/EDS studies. In comparison, there were fewer peaks in the XRD graph of Grím particles which is consistent with previous studies showing that Grímsvötn particles consisted mostly of basaltic glass while Eyjafjallajökull ash was dominated by andesitic glass (Gislason et al., 2011b). No sulfurous compounds were observed in the studies as expected as the samples were collected from the ground in close vicinity of the volcanoes and were not aged in the atmosphere. However, exposure to crystalline silica is well known to causes silicosis (Horwell and Baxter, 2006). In addition, volcanic ash containing Fe2+ can increase the toxic reactions in the lung.

3.2. Properties of aerosolized ash particles

3.2.1. Particle size distributions

The properties of aerosolized ash particles obtained from Eyja and Grím are shown in Table 1 and the corresponding particle number size distributions in Fig. 5. The aerosolization of the ash samples show that both volcanic ashes can be easily re-dispersed into the air. The geometric mean diameter (Dp) of the re-dispersed particles was below 1 μm at the studied flow rates of 15, 20 and 30 L min⁻¹ (Table 1). In the case on Eyja particles, the particle size distributions were unimodal (Fig. 5A–C). The Dp of particles decreased from 830 nm to 669 nm while the total particle concentration was almost tripled as the bed flow increased from 15 L min⁻¹ to 30 L min⁻¹. This is explained by improved de-agglomeration at the higher flow rates. The total number of re-dispersed particles was over 10 000 # cm⁻³. The Dp of the re-dispersed Grím particles was slightly larger compared to Eyja particles, particularly at the flow rate of 15 L min⁻¹. As with the Eyja particles, the size of the Grím particles decreased from 940 nm to 694 nm with increasing flow rate from 15 to 30 L min⁻¹ due to the better de-agglomeration. In contrast to the Eyja particles, the number concentration of particles with diameters around 100 nm increased as the flow rate increased and thus, the particle size distributions became bimodal (Fig. 5D–F). However, the total number concentration of aerosolized particles was around two times lower at the flow rates of 15 and 20 L min⁻¹ and over four times lower at the highest flow rate than those for Eyja (Table 1). This corresponds to the specific surface area measurements and SEM images of the bulk samples (Fig. 3) indicating that ash particles of these volcanoes have different size and shape (rounded vs. faceted) that affected their re-dispersion.

A clearly increase in ground level PM10 values were measured around Europe during the eruptions (Bukowiecki et al., 2011; Colette et al., 2011; Donovan and Oppenheimer, 2011; Schäfer et al., 2011). To compare the ground level measurements, the particle size distributions of re-suspended ash were converted to the mass/volume distributions (one mode log-normal fitting, density 2.0 g cm⁻³). Volume size distributions of re-dispersed particles had a mode around 2 μm (1.7–2.7 μm). A similar mode consisting of volcanic ash particles was also observed at several locations in Europe during the eruptions (Bukowiecki et al., 2011; Kerminen et al., 2011; O’Dowd et al., 2012; Pitz et al., 2011). However, the emission of a large amount of gaseous SO2 to the atmosphere during the eruptions and consequent secondary aerosol formation in the troposphere was not investigated in this study. Thus, the increase in the accumulation mode, i.e. formation of new particles around 0.3–0.5 μm, observed in field studies and other effects of SO2 are not comparable to this study.

3.2.2. Shape and composition of the aerosolized particles

Fig. 6 shows the SEM images of the ash particles aerosolized at 15 L min⁻¹. Similar particle shapes were also observed with the higher flow rates. The particles are significantly smaller than the ash particles collected from the ground (Fig. 3). Furthermore, the re-dispersed particles from both Eyja and Grím showed good resemblance to airborne particles that were collected from the atmosphere during the eruptions in various locations in Europe (Bukowiecki et al., 2011; Kerminen et al., 2011; Petäjä et al., 2012). Based on the TEM/EDS the particles consisted mostly of silicates and iron which is in agreement with the XRD studies of the settled particles.
Table 1
Average particle diameter ($D_p$, geometric mean diameter), geometric standard deviation (GSD), total particle number concentration ($N_{tot}$) and mass concentration (PM$_{10}$) of the Eyjafjallajökull and Grímsvötn ash particles dispersed at different flow rates. PM$_{10}$ values were calculated from log-normal fitted distributions (one mode fitting, density 2.0 g cm$^{-3}$). (± = standard deviation, n.a. = not available).

<table>
<thead>
<tr>
<th>Volcano</th>
<th>Dispersion flow rate [L min$^{-1}$]</th>
<th>$D_p$ [nm]</th>
<th>GSD</th>
<th>$N_{tot}$ [1 cm$^{-3}$]</th>
<th>PM$_{10}$ [mg m$^{-3}$]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Eyjafjallajökull</td>
<td>15</td>
<td>830 ± 8</td>
<td>2.16 ± 0.01</td>
<td>(1.39 ± 0.05) × 10$^4$</td>
<td>(1.14 ± 0.22) × 10$^4$</td>
</tr>
<tr>
<td></td>
<td>20</td>
<td>770 ± 4</td>
<td>2.11 ± 0.00</td>
<td>(2.15 ± 0.06) × 10$^4$</td>
<td>(1.78 ± 0.17) × 10$^4$</td>
</tr>
<tr>
<td></td>
<td>30</td>
<td>669 ± 3</td>
<td>2.11 ± 0.01</td>
<td>(3.88 ± 0.14) × 10$^4$</td>
<td>(3.69 ± 0.18) × 10$^4$</td>
</tr>
<tr>
<td>Grímsvötn</td>
<td>15</td>
<td>940 ± 20</td>
<td>2.15 ± 0.08</td>
<td>(7.95 ± 0.18) × 10$^3$</td>
<td>n.a.</td>
</tr>
<tr>
<td></td>
<td>20</td>
<td>729 ± 6</td>
<td>2.60 ± 0.03</td>
<td>(1.06 ± 0.02) × 10$^4$</td>
<td>n.a.</td>
</tr>
<tr>
<td></td>
<td>30</td>
<td>694 ± 9</td>
<td>2.41 ± 0.03</td>
<td>(8.77 ± 0.39) × 10$^3$</td>
<td>n.a.</td>
</tr>
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</table>

Fig. 5. Aerodynamic number size distributions of Eyja particles aerosolized at flow rates of (A) 15 L min$^{-1}$, (B) 20 L min$^{-1}$ and (C) 30 L min$^{-1}$, and Grim particles aerosolized at flow rates of (D) 15 L min$^{-1}$, (E) 20 L min$^{-1}$ and (F) 30 L min$^{-1}$.
ash samples. The silicate grains are most likely a mixture of various minerals as observed in situ measurements during Eyja eruption (Schumann et al., 2011). The results were consistent with the online measurement data obtained during the eruptions, as particle composition and morphology of the re-dispersed particles were very similar to the results of field experiments. This indicates that the re-dispersed ash particles can be used to evaluate the lung deposition of these particles.

3.3. Lung deposition of the aerosolized particles

Table 2 shows the lung deposited surface area concentrations (LDSA) of submicron ash particles ($D_p < 1 \mu m$) determined with NSAM and ELPI. The results correspond to tracheobronchial (TB) and alveolar (AL) regions of the lung at different experimental conditions (scaled to PM$_{10}$ value of 1 mg m$^{-3}$). The LDSA concentration values indicate the total surface of the particles deposited into the human lung regions per volume of inhaled air. The LDSA values of submicron particles increased with increasing dispersion flow rate from ca. 10–50 $\mu m^2 cm^{-3}$ for the AL region and from ca. 3–10 $\mu m^2 cm^{-3}$ for the TB region. At higher flow rates relatively more submicron particles were dispersed into the air and thus also LDSA values were higher. However, the relative deposition surface area values (% i.e., LDSA values divided by the total surface area of the submicron particles) did not show increase with increasing dispersion flow rates. This indicated that submicron particles were equally dispersed into air. NSAM and ELPI results are in relatively good agreement for TB region, see Table 2. However, the LDSA values obtained for the AL region with NSAM were almost consistently smaller than the values.
Surface area concentrations of submicron particles ($D_{1\mu m}$) ash particles aerosolized at different flow rates. The lung-deposited surface area (LDSA) concentrations were measured with NSAM and calculated from ELPI results ($p < 3$). Percents (%) after $/C_0$ using the ICRP respiratory deposition models for alveolar and tracheobronchial regions (Asbach et al., 2009). To facilitate comparisons, the results were scaled to PM$_{10}$ ($D_{10\mu m}$) concentration of 1 mg m$^{-3}$.

<table>
<thead>
<tr>
<th>Flow rate [L min$^{-1}$]</th>
<th>Volcano Dispersion</th>
<th>Respiratory Deposition, %</th>
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<tr>
<td></td>
<td>AL</td>
<td>TB</td>
</tr>
<tr>
<td></td>
<td>LDSA, NSAM $\mu m^2$ cm$^{-3}$</td>
<td>LDSA, ELPI $\mu m^2$ cm$^{-3}$</td>
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<td></td>
<td>Alveolar</td>
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The deposition of particles with $D_p < 10 \mu m$ into different regions of lungs are shown in Table 3. The results were scaled to a PM$_{10}$ concentration of 1 mg m$^{-3}$ to facilitate comparison of the data. For AL region the deposition fractions of number, surface area and mass were around 10% of the particles in all cases. However, a clear difference was observed for the TB and HA deposition. The average particle deposition fractions for particle number, surface area and mass concentrations obtained at different dispersion flow rates were 1.6%, 3.7% and 4.5% for TB and 15%, 44% and 60% for HA regions, respectively. Compared with number fraction, substantially larger fraction of ash particle surface area and mass can deposit onto TB and HA regions. A rough estimation of the ash particle mass deposited onto the lungs can be made based on our results. The mean daily average PM$_{10}$ value was around 0.5 mg m$^{-3}$ in Reykjavik on June 4, 2010. Assuming 10%, 5% and 60% deposition fractions for the AL, TB and HA regions, respectively, with an average respiratory flow rate of 20 L min$^{-1}$ (1.2 m$^3$ h$^{-1}$), a daily deposition of ash particles onto the AL, TB and HA regions are around 1.4 mg, 0.7 mg and 8.6 mg, respectively (total deposition 10.7 $\mu g$).

The LDSA values give valuable information of the health effects of the particles, since the reactivity of the particles within the lung is nowadays known to be related to the surface area and number of particles more than mass (Horwell and Baxter, 2006). The volcanic ash consisting of fine particles can potentially penetrate deep into the lungs and thus cause coughing and irritation especially to people suffering from lung disorders such as asthma (Horwell et al., 2010). While the immediate respiratory health effect on people living close to the volcanoes could be minimal, the long-term health effect should be monitored (Carlsen et al., 2012). Although a main fraction of surface area is deposited to the upper airways, obtained with ELPI. This discrepancy is most likely caused by the different measurement principles of NSAM and ELPI (Asbach et al., 2009; Keskinen et al., 1992).

Since NSAM can only measure reliably LDSA values for submicron particles, lung deposition of PM$_{10}$ particles ($D_p < 10 \mu m$) were estimated from the ELPI results based on the ICRP respiratory deposition models (Hinds, 1999; ICRP, 1994). Fig. 7 shows a histogram of the particle number size distributions for Grím (flow 30 L min$^{-1}$) compared to the respiratory deposition fraction curves for alveolar (AL), tracheobronchial (TB), head airway (HA) regions and total deposition (Hinds, 1999).
a significant amount of particles can deposit into the alveolar and tracheobronchial regions. In summary, roughly 3–9% and 1–2% of the total surface area of inhaled submicron ash particles was observed to deposit into AL and TB regions of the lungs, respectively. No significant differences between the ashes from Eyjafjallajökull and Grímsvötn were observed. However, the results indicate that a substantial increase in the concentration of respirable airborne ash particles and associated health hazard can take place if the deposited ash particles are re-suspended under dry, windy conditions or by outdoor human activity (Horwell and Baxter, 2006; Thorsteinsson et al., 2012).

4. Conclusions

We have analyzed and compared volcanic ash particles derived near from Eyjafjallajökull (Eyja) and Grímsvötn (Grím) volcanoes during their eruptions. The ash particles were mainly agglomerated and irregular in shape. The Grím particles were larger and more faceted than Eyja particles, which was likely to originate from the different conditions during the particle formation. In both cases, the coarse particles were covered with smaller particles adhered to their surface. Grímsvötn particles consisted of mostly basaltic glass while Eyjafjallajökull ash is dominated by andesitic glass. The specific surface area values of the ash particles were typical for relatively large and non-porous particles. The composition, size, density and morphology of Eyja and Grím ash particles were different, which affects re-dispersion, lung deposition and thus, toxicological properties of the particles.

The aerosolization experiments of the ash samples showed that both volcanic ashes can be easily re-dispersed to the air. The geometric mean diameters of re-dispersed particles were below 1 μm. The results indicate that a substantial increase in concentration of respirable airborne ash particles can take place if the deposited ash particles are re-suspended under dry, windy conditions or by outdoor human activity. This can cause a potential health hazard. The results were consistent with the on-line measurement data obtained during the eruptions of both Eyja and Grím. In addition, particle composition and morphology of re-dispersed particles was similar to field experiments results. This indicates that re-dispersed ash particles can be used to evaluate lung depositions of volcanic ash particles.

Measurements and calculations on lung deposited surface area concentrations indicated that roughly 3–9% and 1–2% of total surface area of submicron ash particles is able to deposit into alveolar and tracheobronchial regions of lungs, respectively. No significant difference between Eyja and Grím ashes was observed. Although, a main fraction of surface area is deposited to the head airways region, a significant amount of particles can be deposited into alveolar and tracheobronchial regions. Based on the estimation of mass deposition of ash particles into lungs, a daily dose is around 1.4 mg, 0.7 mg and 8.6 mg for the alveolar, tracheobronchial and head airways regions (total 10.7 μg), respectively, at PM2.5 concentration of 0.5 mg m–3 (respiratory flow rate of 20 L min–1). However, the surface area of ash particle could be relevant measure when determining the potential risks of the inhaled particles since nearly all the chemistry of the particles undergoes surface reactions.

Acknowledgments

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