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Atmospheric ice nuclei in the Eyjafjallajökull volcanic ash plume

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Abstract

Explosive volcanism affects weather and climate. Primary volcanic ash particles which act as ice nuclei (IN) can modify the phase and properties of cold tropospheric clouds. During the Eyjafjallajökull volcanic eruption we have measured the highest ice nucleus number concentrations (>600 L) in our record of 2 years of daily IN measurements in central Germany. Even in Israel, located about 5000 km away from Iceland, IN were as high as otherwise only during desert dust storms. These measurements are the only ones available on the properties of IN in the Eyjafjallajökull plume. The measured high concentrations and high activation temperature (−8 °C) point to an important impact of volcanic ash on microphysical and radiative properties of clouds through enhanced glaciation.

1 Introduction

The recent eruption of the Eyjafjallajökull volcano in Iceland, besides affecting aviation in Europe, raises the question about potential effects of volcanic emissions on weather and climate. The climate impact of explosive volcanic injections into the stratosphere through the radiative effects of secondary H_2SO_4/H_2O aerosol particles formed from the injected SO_2, is well documented (Robock, 2000). However, only few eruptions reach the stratosphere, and the total emission of 20 Tg yr^{-1} of fine ash into the troposphere by small volcanic eruptions (Mather et al., 2003) exceeds the time-averaged annual return flux of volcanic sulphate from the stratosphere to the upper troposphere by a factor of 10–40. Volcanic ash particles can affect the phase of supercooled tropospheric clouds by acting as ice nuclei (IN) (Durant et al., 2008). About 50% of the global cloud population at −20 °C is found to be supercooled (Choi et al., 2010), i.e. to contain metastable water that freezes upon the presence of suitable ice nuclei. Glaciation affects the phase, size distribution and colloidal stability of the cloud particles, as well as the lifetime, dimensions and radiative forcing of the clouds. Space observations show

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that on the planetary scale the fraction of supercooled clouds (at −20 °C) and the coincident dust aerosol frequency are negatively correlated (Choi et al., 2010), likely due to glaciation by dust. Accompanying model calculations reveal that the cloud albedo is significantly affected upon glaciation.

Despite their relevance, ice nuclei remain an undersampled component of the climate system. A quantitative assessment of their sources and a climatology of their atmospheric abundance are not available. Many components of atmospheric IN have been identified, such as minerals of desert dust, bacteria, pollen and plant debris (Pruppacher and Klett, 1997; Szyrmer and Zawadsky, 1997) although for many of them the concentrations and relevance to cloud processes are still unclear (Möhler et al., 2007; DeMott et al., 2010). In addition to these surface-derived sources, volcanism is debated as a source of atmospheric ice nuclei, with conflicting evidence from field measurements (Isono et al., 1959; Price and Pales, 1963; Hobbs et al., 1971; Schnell and Delany, 1976; Radke et al., 1976). Recent satellite observations (Gasso, 2008) show that natural degassing or weakly explosive volcanoes in the South Atlantic and North Pacific affect low marine stratocumulus for up to 1300 km downwind by decreasing droplet effective radius and increasing visible brightness, and may add cloud cover in otherwise cloudless areas. While the latter observations did not consider explicitly the ice-nucleating ability of the volcanic aerosol, our measurements during the Eyjafjallajökull eruption show a significant enhancement of atmospheric IN when the dispersed ash cloud reached central Europe in April 2010 and the eastern Mediterranean in May 2010.

2 Methods

The number concentration of atmospheric ice nuclei near the surface is monitored at the Taunus Observatory (TO) in central Germany on a daily basis since April 2008 and at Tel Aviv University (TAU) since November 2009 using the same methods and identical instruments. The data discussed here comprise the months of April and May 2010 that were affected by volcanic emissions. The Taunus Observatory is a mountain site

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for atmospheric research located 25 km north of Frankfurt (825 m a.s.l.). The Tel Aviv University sampling site is on the university campus, about 2.5 km from the Mediterranean Sea shore.

Aerosols were collected on 47 mm diameter silicon substrates using a specially designed electrostatic precipitator (Klein et al., 2010a). All the samples were collected at a flow rate of 2 liters min^{-1} with 10 liters and 5 liters sampled at TO and Tel Aviv, respectively. Substrates were analyzed in the isothermal static vapor diffusion chamber FRIDGE (Klein et al., 2010a; Bundke et al., 2008). Ice nuclei concentrations were measured at three different temperatures between −18 °C to −8 °C and RH_{ICE} between 103% and 119%. The ice crystals were observed by a CCD camera and were counted automatically. The regular sampling frequency usually was 1 per day, but was higher during periods of interest.

In those samples that were collected during the volcanically affected days of 16–17 April and 16–17 May the chemical composition and morphology of 150 individual particles that had been previously identified as ice nuclei by FRIDGE were determined by environmental scanning electron microscopy (ESEM) combined with energy-dispersive X-ray microanalysis (EDX).

The unambiguous identification of the analyzed particles as ice nuclei was enabled by a high precision laser engraved coordinate system on the substrates. The positions on the substrates, where ice nucleation was observed by the CCD camera of FRIDGE, can be recovered in the ESEM with a lateral resolution of approximately 5 µm.

The aerosol particle surface area was derived from measurement by an Electrical Aerosol Spectrometer (Tammet et al., 2002) using a particle shape factor of 1.2 and density of 2.6 g cm^{-3}.

3 Results and discussion

In our record of 965 individual daily measurements at TO from April 2008 to March 2010 the ice nucleus number concentrations (at −18 °C and water saturation) range between 1.3 and 312 IN L^{-1}, with a median of 24.2 IN L^{-1}, Klein, 2010; Klein et al.,

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2010b). Figure 1 shows the IN number concentration recorded at TO during the spring of 2010, with the seasonal mean and standard deviation for the months of April–June 2008–2010 indicated by the blue line and vertical bar (pink bands), respectively. The Eyjafjallajökull eruption began on 14 April 2010 and ended on 21 May (Schumann et al., 2010). When the plume was advected over Europe the highest IN peaks obtained so far in our record were detected, which significantly exceed the springtime seasonal mean of $45 \pm 54\,\mathrm{IN\,L^{-1}}$ (arithmetic mean ± standard deviation) at the site. With a high pressure system west of the British Isles the volcanic plume was carried in 1–2 days in a northwesterly descending flow straight across the North Sea towards central Europe where it resided for several days. According to dispersion models, trajectory calculations (Fig. 2), and ceilometer observations of the German Meteorological Sevice DWD the plume reached central Germany in the afternoon of 16 April (Emeis et al., 2010). This was confirmed by sun photometer observations at Leipzig (Ansmann et al., 2010). The IN number concentration at TO began to rise at 15:00 UTC on 16 April, peaked at 10:30 (UTC) on 17 April, and remained high until 21 April. The aerosol lidars at Leipzig and Munich (Ansmann et al., 2010) observed pronounced volcanic ash layers throughout the free troposphere from 16–24 April 2010. The peak of $640\,\mathrm{IN\,L^{-1}}$ on 17 April clearly exceeds the highest peak of $312\,\mathrm{IN\,L^{-1}}$ obtained previously during an intense Sahara dust episode of May 2008 (Klein et al., 2010c). The presence of aged volcanic ash over central Europe during the second half of the month, which was extensively characterized by measurements during many research flights (Schumann et al., 2010) and from ground based Lidar (Flentje et al., 2010), is further reflected in the high monthly mean of $69\,\mathrm{IN\,L^{-1}}$ for April 2010 at TO, as compared to the April means of $46\,\mathrm{IN\,L^{-1}}$ and $18\,\mathrm{IN\,L^{-1}}$ in 2008 and 2009, respectively. High IN concentrations of $100–256\,\mathrm{IN\,L^{-1}}$ were again recorded at TO on 16 and 17 May. For both days the presence of volcanic ash over central Europe is demonstrated by Schumann et al. (2010) from the results of various ash detection systems such as satellite and airborne Lidar measurements and model predictions of the Volcanic Ash Advisory Center.

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In the chemical and morphological analysis of ice nuclei by ESEM four groups of particles were observed: volcanic particles, soot, sea-salt, and biological particles. The volcanic particles were classified on basis of the main and minor elements of these alumosilicates (Mg, Al, Si, K, Ca, Ti and Fe) and/or their glassy morphology. This group can also contain up to 5% (relative abundance) of soil particles, if the monthly average IN concentration in April of previous years is assumed to be present in our volcanic ash samples. The classification criteria for the other groups can be found elsewhere (Ebert et al., 2002). For the samples collected at TO during the peaks of the volcanic ash event on 17 April and 16–17 May the relative number abundance of the different particle groups among the IN are shown in Table 1 (all samples) and Fig. 3 (mean values for each ash event). Confidence intervals (95%) were calculated assuming a multinomial distribution (for details see Weinbruch et al., 2002). The analysis confirms that the volcanic contribution to these particles is at least 63%. As small inclusions of volcanic material were detected within individual sea salt and soot particles the volcanic ash contribution to IN might be even higher.

The signal of volcanic ash in our record of atmospheric IN at TO after the air masses had travelled for at least 2200 km is surprisingly pronounced, in particular when seen in the light of the so far inconclusive and conflicting observational evidence that had been available on volcanic ice nuclei. Isono et al. (1959) found IN at Tokyo, Japan, (at $-20\,^{\circ}\mathrm{C}$) to increase above the 5–20 $\mathrm{IN\,L^{-1}}$ background to up to 50 $\mathrm{IN\,L^{-1}}$ in air that had been affected by Japanese volcanoes located 110–1180 km upwind, whereas others found no evidence for volcanic IN in the effluents of Hawaiian volcanoes (Price and Pales, 1963; Hobbs et al., 1971), of St. Augustine, Alaska (Schnell and Delany, 1976), and of Mt. Baker, Washington (Radke et al., 1976). Some of the latter results were debated recently because the situations sampled were more representative for passively degassing volcanoes rather than for large explosive eruptions and because of a postulated underestimation of IN by the technique applied (Durant et al., 2008; Pruppacher and Klett, 1997).

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A significant volcanic contribution to IN was also recorded a month later at Tel Aviv, more than 5000 km away from the source. Tel Aviv is frequently affected by dust storms, due to the proximity to Arabian and African deserts. During such conditions (defined by a daily average PM_{10} concentration higher than $100\,\mu g\,m^{-3}$; 37 cases), the concentrations of PM_{10} and of IN (mean ± standard error) in 2009/10 were $273.9 \pm 35.5\,\mu g\,m^{-3}$and $53.7 \pm 7.2\,IN\,L^{-1}$, as compared to $41.3 \pm 0.9\,\mu g\,m^{-3}$ and $27.4 \pm 2.0\,IN\,L^{-1}$, respectively, during the "clean" days (with PM_{10} daily average $<60\,\mu g\,m^{-3}$; 109 cases). During 18–23 May 2010, the trajectories (Fig. 2) suggest the advection of air from the north eastern Atlantic around Iceland towards the eastern Mediterranean. While the PM_{10} measurements at TAU during these days indicate "clean" conditions with PM_{10} values well below the $100\,\mu g\,m^{-3}$ level (defined as a dust event), the IN peak at around $100\,L^{-1}$ (see Fig. 3) and the average of $66.9 \pm 37.5\,IN\,L^{-1}$ (5 samples) is significantly higher than the averages for both normal "clean" days and for some "dusty" days. Furthermore, the elemental composition of individual particles that were randomly selected from the samples collected during the passage of the volcanic plume over Israel was analyzed using ESEM-EDX. The analysis shows that the volcanic particles had similar composition to mineral dust, except that sea salt and some sulphate were also present. All these observations clearly point to a strong contribution of volcanic ash to the total IN population over Israel during this event.

Since ice nucleation is considered a surface phenomenon, the specific density of IN number per aerosol surface area is of interest and is important for parameterization used in numerical models of clouds and climate. From our measurements of IN number concentration and aerosol particle size distribution in the 0.3–20 µm diameter size range at TO during the volcanic event we derive a mean IN surface area density of $2 \times 10^9\,IN\,m^{-2}$ of aerosol at −18 °C and water saturation. This is at least a factor of two higher than the IN surface densities that were observed under the same conditions in various tropospheric environments, including air masses affected by mineral dust (Klein et al., 2010c; Phillips et al., 2008).

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In our samples, the first ice formed at −8 °C and water saturation. This is at the upper end of the range of freezing temperatures reported so far for volcanic aerosol (Durant et al., 2008). Many of the previous data, however, were obtained for the immersion and contact freezing modes, whereas our measurements address the deposition and condensation-freezing modes. At a cloud glaciation temperature as high as −8 °C (equivalent to 3.5 km altitude in the Standard Atmosphere) the Eyjafjallajökull volcanic event may have affected even lower tropospheric clouds by glaciogenic seeding. While mid-level stratus normally is considered to play a neutral role in the Earth's radiation budget because it reflects about as much shortwave solar radiation as it absorbs terrestrial radiation (Cotton, 2009), the quantitative effects of massive cloud seeding on both budget entries remains an unresolved question. As was pointed out above, the fraction of supercooled cloud particles within mixed- phase clouds has recently been found to be negatively correlated to the frequencies of dust aerosols (at the −20 °C isotherm) and the glaciation of such supercooled clouds to decrease cloud albedo (Choi et al., 2010). Considering the high effectiveness of the Eyjafjallajökull particles as IN and the large areal extent of marine stratus and altostratus with possible supercooling in the high northern latitudes (Bretherton and Hartmann, 2009; Hartmann, 1994), a large scale natural cloud seeding event may have taken place, with possible consequences for cloud radiative forcing. It is also probable that high concentrations of these particles at higher levels have increased cirrus cloud cover, resulting in changes of radiation fluxes at these levels. Our measurements during the Eyjafjallajökull volcanic event of 2010 show a massive large scale volcanic contribution to the abundance and properties of IN.

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Table 1. Relative number abundances [%] and 95% confidence intervals (in parenthesis) of the different particle groups among IN during volcanic ash events.

Particle group	Sample #905 04/17/2010 ($n = 98$)	Sample #906 04/17/2010 ($n = 51$)	Sample #945 05/16/2010 ($n = 49$)	Sample #948 05/17/2010 ($n = 50$)
Volcanic ash	66.3 (52.2–78.0)	64.7 (45.2–80.3)	81.6 (62.3–92.3)	70.0 (50.2–84.4)
Sea salt	11.2 (5.0–23.1)	13.7 (5.1–32.0)	12.2 (4.2–30.7)	24.0 (11.4–43.6)
Soot	17.3 (9.2–30.3)	7.8 (2.2–24.7)	0.0 (0.0–13.8)	2.0 (0.2–16.8)
Biological	5.1 (1.6–15.3)	13.7 (5.1–32.0)	6.1 (1.4–22.9)	4.0 (0.7–19.7)

n = total number of particles.

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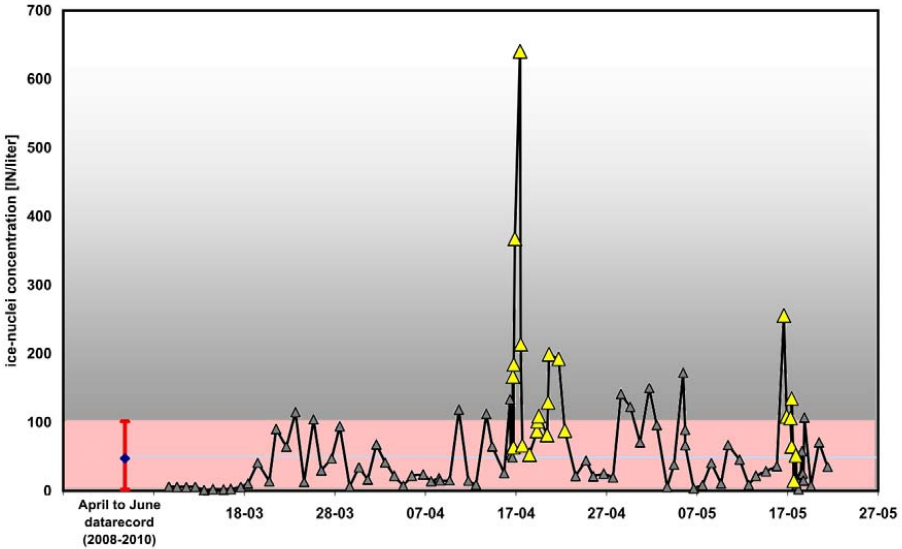

Fig. 1. Number concentration of atmospheric ice nuclei (at −18 °C and water saturation) at the Taunus Observatory (TO) in central Germany during April–May 2010. The blue line and pink banded area are the seasonal average and standard deviation, respectively, for the months of April through June 2008–2010.

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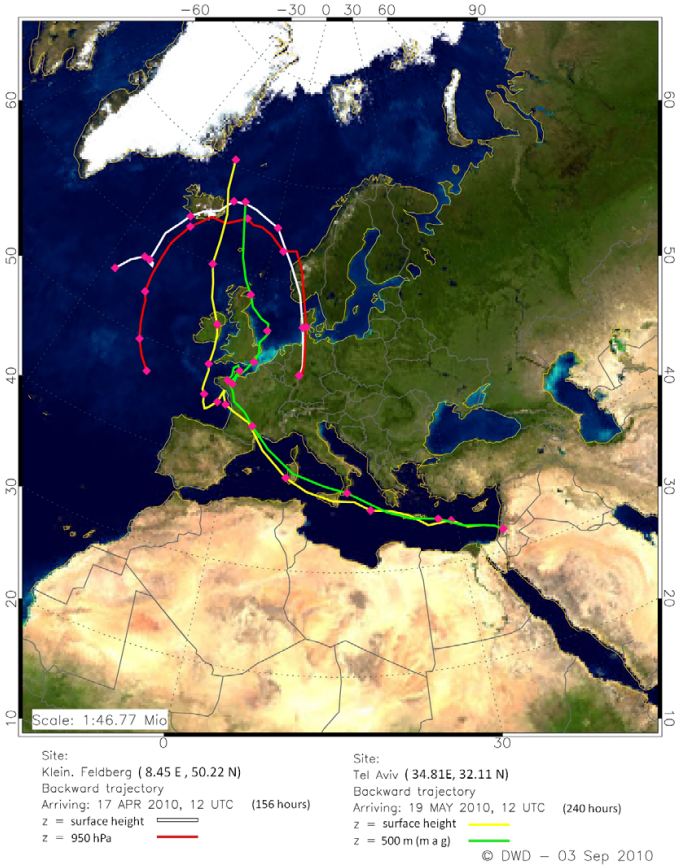

Fig. 2. Backward trajectories from Eyjafjallajökull volcano arriving at Taunus Observatory in April 2010 and at Tel Aviv in May 2010. Trajectories were calculated by the German Weather Service DWD.

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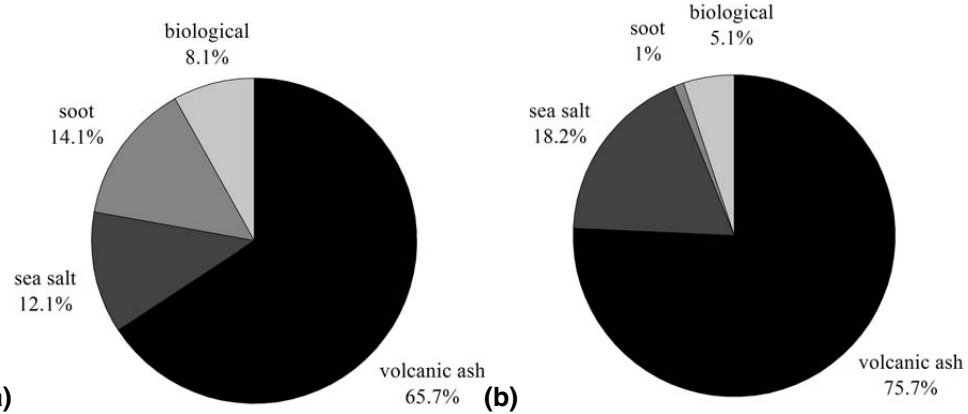

Fig. 3. Average relative number abundance of different particle groups among ice nuclei at TO during volcanic ash events: **(a)** April (samples #905 and #906) and **(b)** May (samples #945 and #948).

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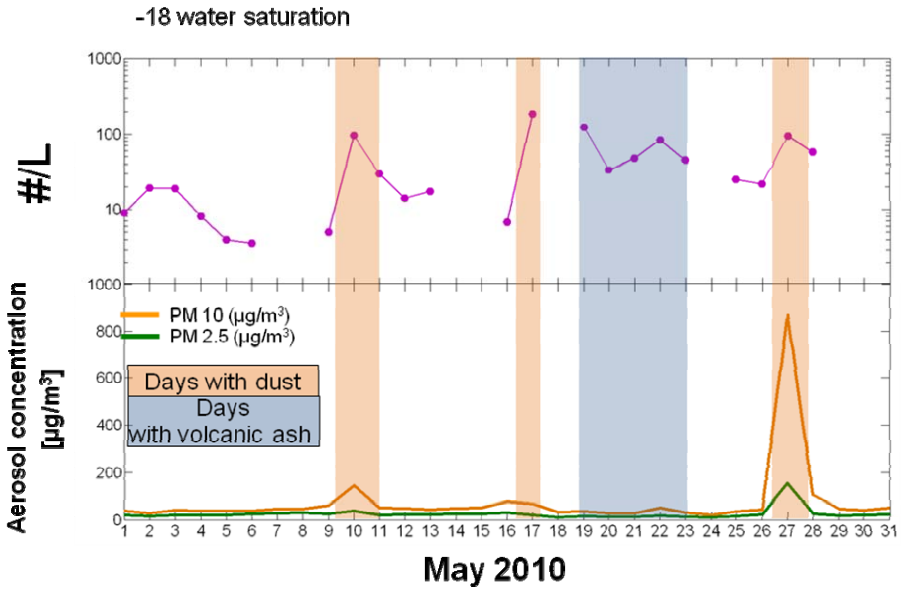

Fig. 4. Number concentration of ice nuclei (at −18 °C and water saturation) and concentration of particulate matter below 10 µm diameter (PM_{10}) and below 2.5 µm ($PM_{2.5}$) at Tel Aviv University during May 2010.

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