Raman Microspectriscopic Identification and Characterization of Individual Airborne Volcanic Ash Particles

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Volcanic eruptions can have significant impact on climate, public health and air traffic safety. During the eruption of the volcano Eyjafjalla in Iceland (63.63° N, 19.62° W, 1666 m a.s.l) in April/May 2010, volcanic ash (VA) plumes were ejected into the atmosphere up to 9 km a.s.l. (Sigmundsson *et al.* 2010) and distributed over Europe, which ceased air traffic in 23 European countries for several weeks. Different methods for the analysis of chemical composition and structure of VA were applied during the last three years (Gasteiger *et al.* 2011; Gislason *et al.* 2011; Schumann *et al.* 2011; Weber *et al.* 2012), however, to our best knowledge, no work concerning identification and/or characterization of individual airborne VA particles from Eyjafjalla (or from other volcanoes) using Raman Microspectroscopy (RM) was published. RM, which is based on the effect of an inelastic light scattering by molecules, provides fingerprint spectra and allows non-destructive distinction of a wide range of chemical substances in air particulate matter with the spatial resolution of an optical microscope (Ivleva *et al.* 2007).

Samples of fresh and aged airborne VA particles suitable for single-particle RM analysis were collected during aircraft research flights (Schumann *et al.* 2011), operated by "Deutsches Zentrum für Luft- und Raumfahrt" (DLR) on May 2nd (from Keflavik to Stornoway; top part of the fresh plume over the North Atlantic) and May 18th (aged ash plume survey over Germany and North Sea). In addition, aerosol samples were collected at ground level in an urban setting at the Institute of Hydrochemistry in Munich (TUM), using the Electrical Low Pressure Impactor (ELPI). Furthermore, volcanic ash particles taken at the ground near the eruption (June, 2010) and basaltic rock were studied by RM. Particle size and morphology, Raman patterns (revealing mineral composition and structure), and the particle stability with respect to the laser beam intensity were taken into account. We analyzed similarities between Eyjafjalla VA particles, collected at the different sampling sites, and compared them with the spectra of more than forty glassy and crystalline minerals (SpecID, Horiba Scientific) by applying cluster analysis (CA), in order to achieve information on structure and composition of volcanic ash.

Figure 1 shows representative VA particles from fresh (fp) and aged (ap) plumes, the corresponding Raman spectra, and spectra of some reference compounds. The fp and ap VA particles are brownish and can change color and shape when they melt in the focus of the intense excitation laser beam. The particles exhibit similar spectral pattern with relatively broad bands near 310, 390, 415, 540, and 720 cm^{-1}, suggesting the presence of different minerals with rather disordered crystalline structure (Ivleva *et al.* 2013). A very broad band, which extends from ca. 200 cm^{-1} to ca. 700 cm^{-1}, is typical for glassy structures, such as SiO_2 glasses. We can assume a complex structure of VA particles, where different mineral phases are embedded in glassy matrix. Slight differences in the spectra of fresh and aged VA particles suggest variable proportions of the different minerals, partial precipitation, or chemical aging of VA during the transport. In addition, soot, sodium nitrate and titanium dioxide particles were found in aged plumes and particular matter sampled on ground level, indicating anthropogenic influence. These results show that RM is indeed an effective method for identification and chemical characterization of individual airborne volcanic particles.

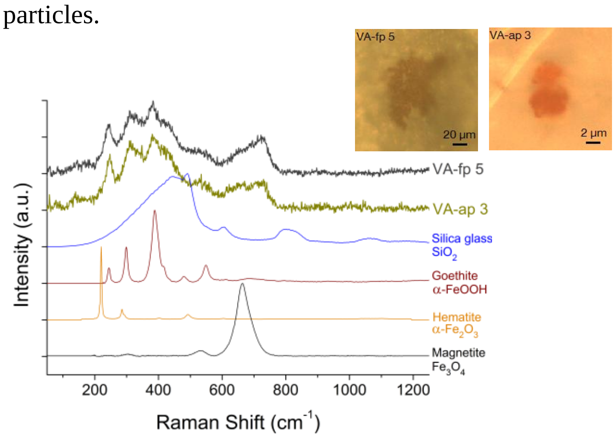

Figure 1. Optical microscope images of VA particles from fresh (fp) and aged (ap) plumes, corresponding spectra of VA and reference compounds.

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