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# A new method for sampling of atmospheric ice nuclei with subsequent analysis in a static diffusion chamber

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ABSTRACT

A new technique for the sampling of atmospheric ice nuclei is presented. Aerosols are electrostatically precipitated onto silicon discs, and are subsequently analyzed in a diffusion chamber. The method is compared to continuous flow chamber measurements and to membrane filter measurements of ice nuclei. Ice nucleus measurements obtained using this new method compare well to the continuous flow chamber measurements. In contrast, membrane filters were found to lead to serious underestimates of the ice nuclei number concentration when the filters were treated with a pore sealant and analyzed under vacuum. This confirms previous findings of other authors.
The electrostatic aerosol collection device is lightweight and easy to handle.


## 1. Introduction

Aerosols which are active in clouds act as Cloud Condensation Nuclei (CCN) or Ice Nuclei (IN), and are an essential ingredient for the formation of tropospheric clouds, affecting both the formation and the properties of clouds. The cloud albedo effect of anthropogenic aerosols on radiative forcing in the global climate system has been assigned by far the highest uncertainty of all anthropogenic perturbations, and a low level of scientific understanding (IPCC, 2007). Measurements of these aerosols still remain scarce, and we are still far from a climatological view on these atmospheric constituents.

Ice nuclei are defined operationally as those atmospheric aerosol particles that generate ice on their surface when the aerosol is exposed to sub-freezing temperatures and supersaturation with water vapor. Such conditions are provided for the measurement of IN in continuous flow chambers (CFC) (Rogers et al., 2001; Hussain and Saunders, 1984; Bundke et al., 2008; Stetzer et al., 2008), as well as in chambers that expose a substrate laden with aerosol particles from the atmosphere (Bigg et al., 1963; Stevenson, 1968; Bigg, 1990) to such conditions. The ice crystals that grow on the nuclei are subsequently counted. These methods usually address the deposition freezing mode (and the condensation freezing mode close to water saturation). Continuous flow techniques have a high time resolution compared to off-line techniques, but at the expense of a high experimental and logistical effort for field measurements. In their most advanced form these techniques are coupled to spectrometers for the analysis of the composition of individual particles (Kreidenweis et al., 1998; Cziczo et al., 2003; Kamphus et al., 2007). If high time resolution is not required, such as in ground-based and monitoring activities, off-line techniques may be more advantageous. A method that was first proposed by Bigg et al. (1963) and which has been widely applied is the collection of aerosols on membrane filters (in the following referred to as "the filter method"), followed by the activation of ice nuclei in a diffusion chamber and the counting of the ice crystals grown. Various different types of processing chambers have been used (Bigg et al., 1963; Langer and Rodgers, 1975; Meyer and Gravenhorst, 1976; Stein and Georgii, 1985; Fountain and Othake, 1985; Hussain and Kayani, 1988). The method is easy to handle, but can be affected by a potential depletion of water vapor through hygroscopic particles on the substrate, which may suppress the "activation" of some ice nuclei. This artifact is known as the "volume effect",

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as it appears to be proportional to the volume of air sampled. Among several proposed measures to overcome this effect (Huffman and Vali, 1973; Lala and Jiusto, 1972; Langer and Rodgers, 1975; Bigg, 1990) Gravenhorst et al. (1975) reduced the pressure in their diffusion chamber to enhance the diffusion of water vapor and thus counteract the vapor depletion.

Questions regarding the efficiency of the filter method have been raised as a result of comparisons between the filter method and continuous flow systems. Hussain and Kayani (1988) found that (at water saturation) their CFC detected a factor of $14 \pm 4$ more ice nuclei than their static diffusion chamber. Archuleta (2002) compared results of measurements over the Arctic Ocean using the Colorado State University's continuous flow diffusion chamber (Rogers, 1988) to filter samples that were collected in parallel to the measurements and that were analyzed by the dynamic filter processing chamber of CSU (Langer and Rodgers, 1975). She concluded "that filter processing severely underestimates the number of ice nuclei", on average by a factor of 33 (Archuleta, 2002).

We have experienced similar deficiencies in the filter method in our own laboratory, which led us to develop a new method for the electrostatic precipitation of ice nucleating aerosols on electrically conductive solid substrates. We have built a vacuum diffusion chamber to activate and count ice nuclei on these substrates using state of the art technology. This chamber, which was briefly introduced by Bundke et al. (2008), is described in more detail in chapter 2.1. The new electrostatic aerosol collection method is described in chapter 2.2, and is intercompared to continuous flow chamber IN measurements in chapter 3.1. Chapter 3.2 reevaluates the filter method by comparing it to both the new method and to the continuous flow IN measurements.

## 2. Experimental

### *2.1. The detection of ice nuclei in the static vapor diffusion chamber FRIDGE*

A static vacuum vapor diffusion chamber, FRIDGE (Frankfurt Ice Nuclei Deposition Freezing Experiment, Bundke et al., 2008), was built for the activation, growth and counting of ice nuclei collected on substrates. It is based on the design by Meyer and Gravenhorst (1976), but uses state of the art equipment. This device can be used for the analysis of ice nuclei collected on any type of substrate of up to 50 mm diameter.

The cylindrical body of the chamber (brass, inner diameter: 70 mm, height: 30 mm) has separate top and bottom halves (Fig. 1) which are sealed together by O-rings. Both parts are mounted on a slide rail, allowing them to be separated vertically. Thus the chamber can be opened to allow substrates to be placed on the central Peltier-cooled plate in the lower part of the chamber, or to be removed. The top part of the chamber is connected to an external water vapor source, to a vacuum pump (Leybold turbomolecular TP50), and to pressure gauges (Datametrics barocell). The water vapor source is an 800 ml cylindrical, thermostatically controlled stainless steel vessel containing ice. The growth of ice crystals on the substrate is observed by a CCD camera (Allied Oscar F-510, $2588 \times 1958$ pixels, magnification: 13.6) through the glass-window on top of the chamber. The sample is illuminated by an LED-ring (Schott, VISILED S40-55, 40

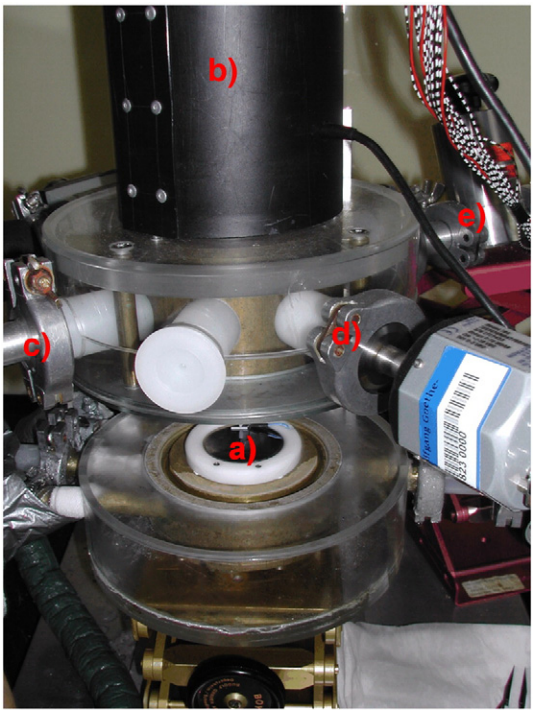


**Fig. 1.** Photograph of the vacuum diffusion chamber FRIDGE while it is open to allow the exchange of samples. The cooled table for the substrate (a) is in the lower part, and the CCD camera (b), connectors to the water vapor source (c), pressure measurement (d) and vacuum pump (e) in the upper part.

High Brightness LEDs, color-temperature 5600 K). The surface temperature of the substrate is measured by a micro PT1000 sensor on the substrate surface, and is directly controlled in the range of 0 °C to −35 °C by adjusting the cooling rate of the Peltier element via a PID controller. The temperature varies by less than 0.04 °C (at −13 °C) over the entire surface, when silicon discs (described in chapter 2.2) are used as a substrate. The mean ± standard deviation of the temperature measurements at the center and at two locations near the edges of the disc were: $-12.98 \pm 0.023$ °C, $-13.01 \pm 0.036$ °C and $-12.98 \pm 0.037$ °C, respectively. All instrumental parameters are fully controlled via LABVIEW™.

Samples are processed as follows: A substrate laden with aerosol is placed on the filter table. The surface temperature sensor is placed near the edge of the substrate. The chamber is closed, evacuated (1 min), and cooled to the desired temperature (2 min). A reference picture of the substrate is taken prior to activation of the nuclei and ice crystal growth. This picture is later subtracted from the images recorded after activation, in order to eliminate inhomogeneities of the substrate background. Water vapor is now introduced by opening the connection to the vapor source. The resulting pressure in the chamber only depends on the temperature of the vapor source. The water vapor supersaturation over the substrate surface is calculated from the pressure in the chamber and the temperature of the surface. The water vapor pressure is measured by a Datametrics barocell pressure sensor and is controlled (PID) by adjusting the temperature of the water vapor source. Thus it is possible to adjust the substrate temperature and the corresponding supersaturation