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A Double Inversion: Size and Time Resolved Growth Rates for Aerosol Particles in the CERN CLOUD Experiment

C. Williamson[a], J. Curtius[a], A. Kürten[a], J. Almeida[a], S. Ehrhart[a], D. Wimmer[a,b], K. Lehtipalo[b], J. Tröstl[c] and the CLOUD collaboration.

[a] *Insitut für Atmosphäre und Umwelt , Goethe University of Frankfurt, Frankfurt am Main, Germany*
[b] *Department of Physics, University of Helsinki, Helsinki, Finland*
[c] *Paul Scherrer Institut, Laboratory of Atmospheric Chemistry, Villigen, Switzerland*

Abstract. An integrated analysis of multiple instruments measuring concentrations of aerosol particles in the CLOUD (Cosmics Leaving OUtdoor Droplets) chamber at CERN obtains size and time dependent growth rates. A matrix inversion is performed twice: first to get size distributions from measured concentrations using instrument transfer functions; secondly to get growth rates from size distributions using the aerosol general dynamic equation (GDE).

Keywords: Aerosols, Growth Rates, Chamber Experiments, Inversion Methods
PACS: 92.60.Mt

THE CLOUD-EXPERIMENT

Ion-induced nucleation is one mechanism proposed to explain a possible link between galactic cosmic rays (GCRs) and cloud formation [1]. Along with the role of ternary species, this can be better understood through analysis of aerosol particle growth rates under controlled conditions. CLOUD aims to perform this analysis using an aerosol chamber containing synthetic air, controlled levels of sulfuric acid, amines and organics [2].

Data was collected over a two month measurement campaign in 2012. Aerosol nucleation and growth rates were investigated under various conditions including: ternary nucleation with ammonia/DMA, pure ozonolysis and pure OH oxidation of α-pinene and simulation of atmospheric conditions at SMEAR II [3]. Neutral, GCR and CERN-PS beam conditions were used to study ion effects. Many instruments continuously measured particle concentrations in the chamber, including: two

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diethelene-glycol condensation particle counters (DEG-CPCs)[4], a scanning and a fixed particle-size-magnifier (PSM)[5], a CPC-battery (TSI CPCs 3776, 3772, 3010), a laminar diffusion tube (LDT)[6] and a scanning mobility particle sizer (SMPS)[7].

THE INVERSION

Inversion 1: from Concentrations to Size Distributions

Each instrument or instrument channel measures particles of varying sizes with varying counting efficiencies, characterized by transfer functions as shown in figure 1. These transfer functions, $f_i\left(D_p\right)$, relate the measured concentrations, b, to the size distribution, $\frac{dN(D_p)}{d\log D_p}$, as follows:

$$f_i(D_p)\,\frac{dN(D_p)}{d\log D_p}\,d\log D_p = b_i \qquad \text{Eq. 1}$$

Eq.1 is discretized giving a matrix, **A**, of the transfer functions, whose pseudo-inverse is found using singular value decomposition. Multiplying this with the vector of measured concentrations, $\vec{b}$, gives a set of possible size distributions, $\vec{x}$:

$$\vec{x} = \boldsymbol{A}^{-1}\,\vec{b} + \boldsymbol{V}\,\vec{a} \qquad \text{Eq. 2}$$

where $\boldsymbol{V}$ is the null-space of $\boldsymbol{A}$ and $\vec{a}$ is an arbitrary vector.

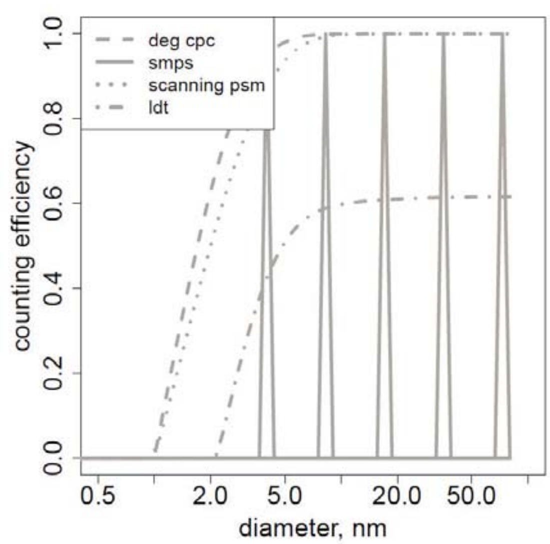

FIGURE 1. Transfer functions from instruments measuring particle concentrations in the CLOUD chamber. Only selected channels are shown , namely DEG CPC 1, SMPS channels 4.0 ,8.3,17.1,35.5,55.0 nm, scanning PSM 0.16lpm flow, LDT 1550cm3/min flow.

A solution is found assuming nothing about the functional form of the solution except that the size distribution is non-negative and smooth, correcting for inherent bias to larger size bins and weighted each measurement according to its uncertainty [8].

Before application to data from CLOUD, this method was tested on simulated size distributions. Expected instrument responses for each size distribution were calculated using transfer functions, varied within instrumental uncertainty and run through the inversion method. The input and inverted size distributions can then be compared as shown in figure 2.

Inversion 2: from Size Distributions to Growth Rates

The GDE for aerosol particles can be expressed as follows [9]:

$$\frac{dN_\Delta(t)}{dt} = \frac{dN_\Delta}{dD_p}\left(D_{p1}, t\right)\frac{dD_{p1}}{dt} - \frac{dN_\Delta}{dD_p}\left(D_{p2}, t\right)\frac{dD_{p2}}{dt} + C_\Delta\left(D_{p1}, D_{p2}, t\right) \qquad \text{Eq. 3}$$

Where Δ is the size interval between particle diameters D_{p1} and D_{p2} and $\frac{dD_p}{dt}$ is the growth rate. The combined coagulation sources and sinks for each interval (C_Δ) are calculated using the Fuchs coagulation rate coefficient [10].

Eq. 3 can be discretized and rearranged to take on the following vector form

$$\frac{dN_\Delta}{dD_p}\overrightarrow{GR} = \overrightarrow{\frac{dN_\Delta(t)}{dt} - C_\Delta} \qquad \text{Eq. 4}$$

This is inverted in the same way as eq. 2 to retrieve the vector of growth rates, $\overrightarrow{GR}$.

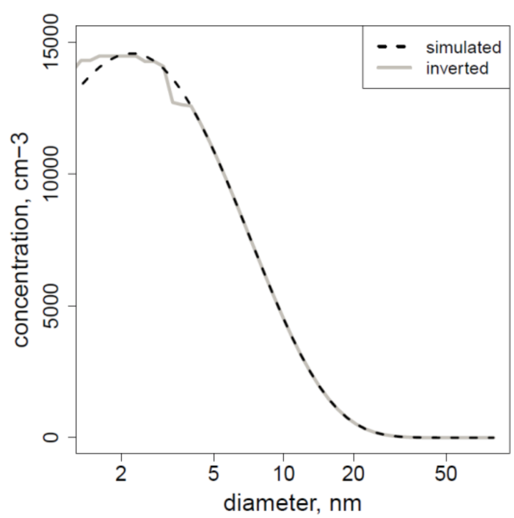

FIGURE 2. A simulated size distribution (dashed black) and an inversion of predicted concentrations obtained from this distribution (solid grey).

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