Hygroscopic properties of large aerosol particles using the example of aged Saharan mineral dust – a semi-automated electron microscopy approach

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Introduction

Mineral dust is regularly transported from the Saharan desert across the Atlantic Ocean to the Caribbean (Ridley et al. 2012). During their transport, dust particles may undergo aging and mix sea-salt or secondary aerosol particles like sulfates or organics. This aging alters the hygroscopic behavior like growth factor and deliquescence relative humidity and therefore affects the physical and chemical properties of the aerosol. Furthermore, in the Caribbean large quantities of soluble particles are admixed to the aged desert aerosol.

From June 10 to July 15 2013, the Saharan Aerosol Long-range Transport and Aerosol-Cloud-Interaction Experiment (SALTRACE) took place. During that period aerosol samples were collected ground-based at Ragged Point, Barbados (13°9'54.4"N, 59°25'55.7"W)

Growth factor

Methodology

- with Atomic Force Microscopy (AFM) the 3-dimensional shape of some drops at various relative humidities were measured
- contact angles and volume of the drops were determined
- with that an empirical formula for the correlation between drop diameter D_{drop} and volume V_{drop} was derived :

$$V_{drop} = 0.5185 \cdot D_{drop}^{2.0677}$$

- the growth factors (Fig. 2) were calculated by: $\sqrt[3]{\frac{V_{drop}}{V_{particle}}}$
- most abundant particle classes are sea-salt (low Cl-S index) and soluble sulfates
- the Cl-S index, defined as S/(S+Cl), was introduced as an simple indicator for how much aging the particle underwent

Results

- the gap between the theoretical growth factors and the observed ones in Fig. 2 shows that aging affects the growth factor indeed
- silicates formed droplets, but it was only a small fraction (<5%) of the total amount of silicate particles and only at high relative humidity. This behviour is corroborated by the chemical analyses, showing that only a minor fraction of the dust particle are internally mixed with considerable amounts of sulfate or sea-salt
- Further investigation of aircraft samples directly out of the Saharan Aerosol Layer (SAL) will show if the silicates hygroscopical behaviour is due to impurities (like sea-salt, sulfate, etc.) the particle has took up in the Planetary Boundary Layer (PBL)

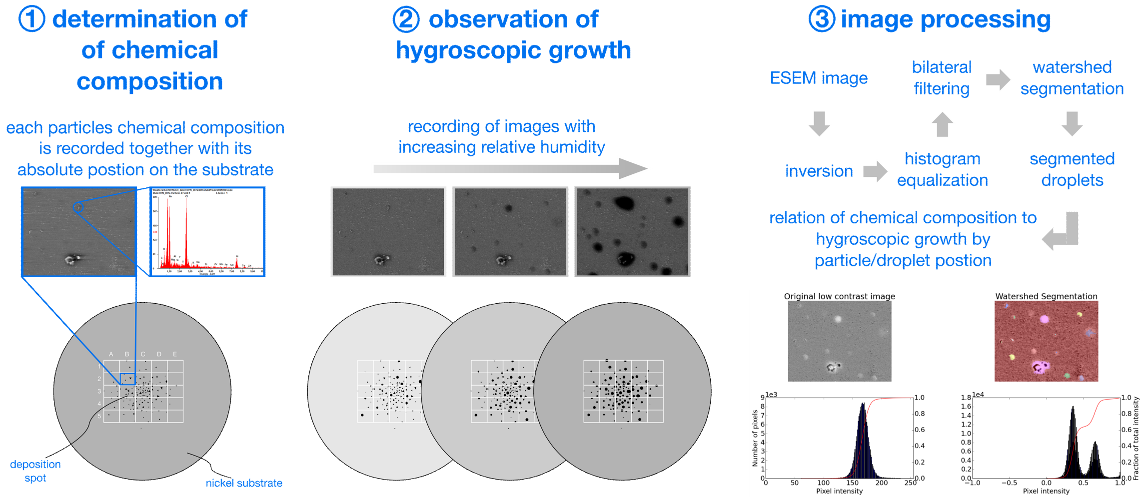

Figure 1. Schematic of the necessary steps for data acquisition.

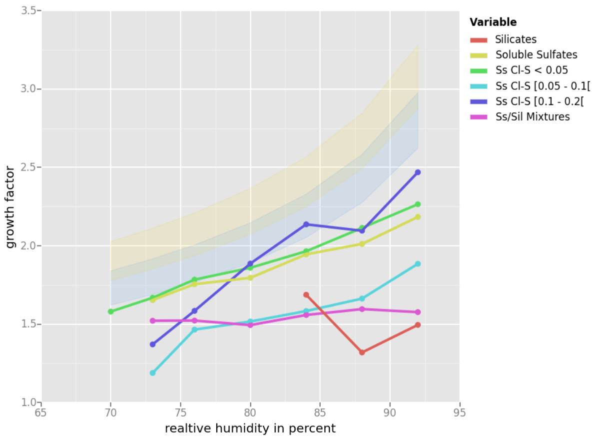

Figure 2. Growth factors for different classes. The semi-transparent areas indicate the range of the theoretical growth factors for NaCl (blue) and sulfate species (yellow) derived from Petters et al. (2007) and references within. At lower humidities, where no curve is shown, hygroscopic growth could not be observed. Ss = sea-salt, Cl-S = chlorine-sulfur index, Sil = silicate. For reasons of clarity nor error bars are shown.

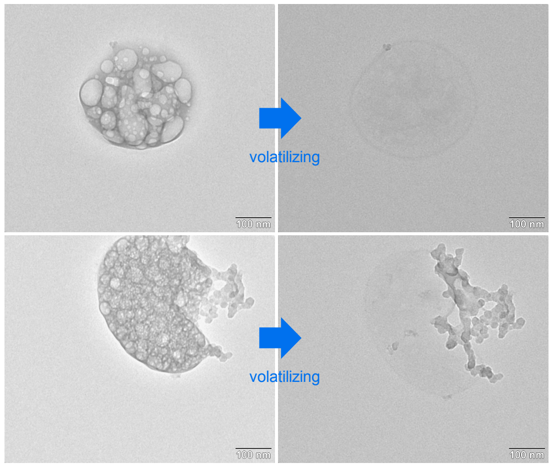

Figure 3. Example of TEM bright field images of two particles before (left) and after (right) after their volatile fraction was evaporated.

Sampling and Analysis

Particle collection

- single round jet cascade impactor
- 50% efficiency cutoff size of approximately 1.1 µm
- Nickel substrate (good thermal conductivity & no interference with measured elements)
- collection time between 10 and 45 minutes depending on dust situation

Analysis

1. „Chemical" run

- automated environmental scanning electron microscopy with energy dispersive X-ray spectroscopy.
- environmental parameters: temperature T = 20°C; H_2O partial pressure e = 1 mbar
- 25 fields of size 73.79 µm x 57.76 µm were analysed on each sample
- quantification of single particle chemical composition
- classification according to chemical composition

2. Image run

- relative humidity (RH) is increased in steps (e = 4.5 mbar = const.; T = decreasing)
- 12 RH-steps between 50 %RH and 92 %RH
- After every RH increase electron microscope images of the exact same fields as in the „chemical" run are recorded

3. Image processing and drop parameter acquisition

- every image undergoes various image processing steps, which lead to segmented single droplets
- from the segmented droplets their respective diameter is derived
- by matching the position of the particle and the droplet, to each individual particle the chemical composition and the size of the respective droplet at a certain RH value can be assigned
- the image processing and the matching of particle and droplet data was automated in a Python algorithm

Refractory fraction

- For sub 500 nm particles, the hygroscopic growth method could not yet be applied. Instead, we show single particle analyses of the refractory / non-refractory fraction here, which supposedly will yield also information on the soluble volume of the particles.
- the high energy electron beam of an Transmission Electron Microscope (TEM) was used to evaporate the volatile fraction of small sulfate particles (Fig.3)
- sample also taken at Ragged Point during SALTRACE 2013
- about 1/3 of all 157 in this way analysed particles have inclusions
- 12% of the particles have inclusions larger than 10% of the whole particle
- the refractory components of internal mixtures with sulfate are mostly soot

References

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Ridley, D. A., C. L. Heald, and B. Ford. 2012. "North African Dust Export and Deposition: A Satellite and Model Perspective." Journal of Geophysical Research: Atmospheres 117 (2): 1–21.

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