# Winter Aerosol in a Mountainous Region of Northern Bavaria

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## INTRODUCTION

Ammonia, nitric acid, nitrogen oxides, and the ammonium and nitrate ions contribute essentially to atmospheric input of nitrogen into the ecosystem. We are interested in their sources, phase distribution, and deposition patterns. Only with knowledge about single particle composition we will be able to understand the microphysical processes at the atmosphere–vegetation interface. We present results from a winter field campaign in a mountainous region of Northern Bavaria (**Fig. 1**).

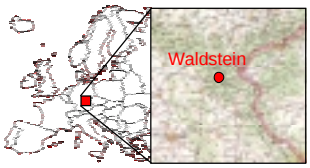


**Fig. 1**: Map of Europe and NE-Bavaria

**Fig. 2**: The »Waldstein« sampling site

## METHODS

Ambient airborne particles were sampled at our ecosystem research site in the Fichtelgebirge mountains (765 m asl, 50°08'40"N, 11°51'55"E; **Fig. 2**) from February 1 through February 18, 2000. We used a transportable time-of-flight mass spectrometer (Trimborn *et al.*, 2000) for on-line analysis of chemical composition of single aerosol particles. Particles were sampled in 6 different size classes ranging from 0.2 to 1.5 µm diameter for 10 minutes each hour. Typical aerosol population patterns were obtained by evaluating all particles of a certain day and size class by fuzzy-cluster analysis according to Hinz *et al.* (1999). For quantitative measurements, a 5-stage impactor was used as a reference sampler for sulfate, nitrate and ammonium ion concentrations of aerosol particles ranging from 0.05 to 10µm aerodynamic diameter.

## RESULTS

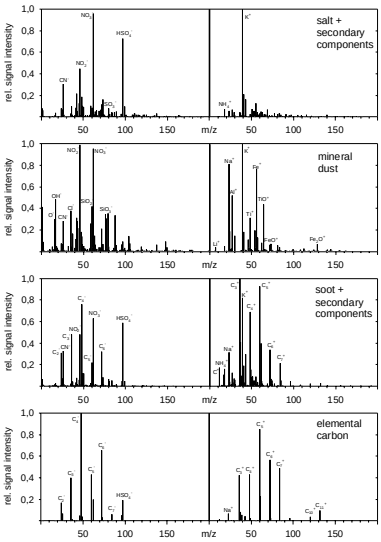


**Fig. 3**: Examples of chemical particle classes

We could distinguish several chemical classes **(Fig. 3).** The classification was performed independently for each day of measurement and particles size, and lead to similar results.

The quality of classification, as evaluated from membership coefficients of fuzzy-clustering of single particle spectra, was found to be good. Over the entire sampling period, membership coefficients of at least 0.8 were assigned to 65 to 80 % of single particle spectra.

Single particle spectra $NO_3^-/NH_4^+$ signal intensity ratios were averaged in order to make them comparable to impactor data ($NO_3^-/NH_4^+$ ratios) to the maximum possible degree (Monte-Carlo-type simulation). Single particles of a unique population were chosen randomly one after another, and combined as random sub-samples of the particle population. After addition of a new particle, the $NO_3^-/NH_4^+$ ratio was re-evaluated by averaging information of the particles so far added to the sample. By consecutively repeating this procedure, Gaussian distributions of mean $NO_3^-/NH_4^+$ ratios for the respective sub-samples of randomly chosen single particles were obtained. **Fig. 4** gives an impression of the changing distribution patterns as more and more particles are added to the sub-sample. The vertical axis represents the relative probability of $NO_3^-/NH_4^+$ ratios for a given number of evaluated particles. Red lines indicate the 95% confidence interval of the Gaussian distribution, the blue line indicates distribution modes.

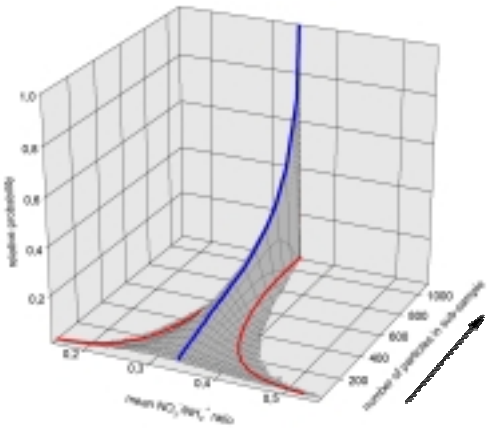


**Fig.4**: 3D representation of Gaussian distributions of $NO_3^-/NH_4^+$ ratios for particle sub-samples

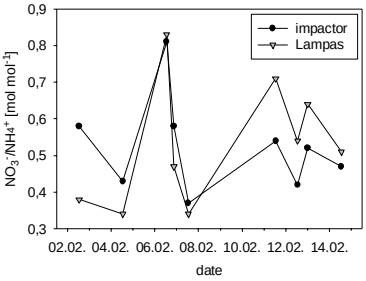


**Fig. 5**: $NO_3^-/NH_4^+$ ratios during our field campaign

**Fig. 5** shows a comparison of the particulate $NO_3^-/NH_4^+$ ratio determined by single particle analysis *versus* impactor measurements. The single particle ratios showed significant scatter due to analytical variability (LDI) and most probably also due to particle-to-particle variations in chemical composition (external mixture). Averaging single particle ratios of a sampling period and using relative sensitivity factors yields reasonably good agreement between single particle ratios and quantitative impactor measurements.

## Conclusions

Clear identification of distinctive chemical classes indicates external mixing of the atmospheric aerosol at our site.

Impactor measurements do not account for the observed particle-to-particle variability. They provide bulk information on atmospheric aerosol properties.

However, the variation of single particle chemical composition has to be considered for a better understanding of phase partitioning and deposition processes of nitrogen species ($NH_4^+$ and $NO_3^-$). This information may be obtained through single particle analysis.

REFERENCES

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