Remote Sensing of the Earth's Atmosphere by Means of Infrared Spectroscopy: Retrieval Theory and Application (Abstract)

Remote sensing retrieval theory covers the methodology used to infer information on unknown parameters of the object under investigation from indirect remote measurements. Application of infrared spectrometry of the atmosphere leads to the inverse solution of the atmospheric radiative transfer equation. Its successful inversion depends on (1) a suitable forward model for simulation of atmospheric radiative transfer (2) the appropiate definition of the retrieval parameter vector, (3) an optimized vector containing the measurements, (4) the appropriate formulation of a constraint, and (5) a tool for stable numeric inversion. Experiments with limb emission spectrometers as well as uplooking emission and absorption spectrometers serve as examples for discussion of application of relevant retrieval schemes.

With respect to retrieval theory, the following progress is reported and the following findings were gained:

- An optimized radiative transfer model for application within a retrieval program was developed. This model is both accurate and efficient. It includes all radiative processes relevant to application to atmospheric infrared spectrometry, including refraction, absorption, emission, scattering, line-coupling and non-local thermodynamic equilibrium.

- Objective selection of spectral gridpoints used for data analysis reduces the retrieval error and reduces computer resources needed. The more spectral data are considered, the more information is used for the retrieval of the target parameter. This is counterbalanced by the fact that radiance at the considered gridpoints depend on further parameters which may be uncertain and thus contribute to the error budget. Tentative retrieval of all these parameters from the measurement leads to an impracticable large number of unknowns. Therefore, an objective method for an optimized selection of spectral data was developed.

- It was shown that for limb measurements of medium spectral resolution the unconstrained solution of the inverse problem is instable if the vertical profile of the target parameter is represented on a discrete level model instead of a discrete layer model. Regularization is needed for level models, while retrieval of parameters represented by layers typically are stable.

- Solvability of ill-posed inversion problems in the context of remote sensing retrieval problems by implicit or explicit regularization was demonstrated. For explicit regularization in particular a smoothing constraint was investigated and considered useful. Implicit regularization we understand is the reduction of the size of the parameter

vector by appropriate discrete representation of vertical profiles of atmospheric state parameters.

- It was shown that the global fit analysis of a limb sequence of spectra is not at all always superior over sequential onion peeling analysis, as commonly believed. In particular in the case of medium spectral resolution measurements the advantage of gobal fit with respect to accuracy disappears and the efficiency of onion peeling becomes significant.

Methods developed were applied to the following experiments:

- The MIPAS (*Michelson Interferometer for Passive Atmospheric Sounding*) balloon-borne instrument; these were the first high spectral resolution limb emission measurements in a winter polar vortex. Vertical profiles of some trace gases relevant to ozone research were retrieved under polar night conditions for the first time.

- The MIPAS aircraft instrument; the MIPAS–FT (Aircraft Transall) instrument recorded the first scientifically useful high resolution spectroscopic measurements under positive elevation angles. These allowed retrieval of zenith column amounts of atmospheric constituents sampled along the flight trajectory independent of solar illumination.

- The ground based MIPAS instrument; The MIPAS–LM (Laboratory model) was used for measurement of time series of various stratospheric trace gases in polar winter by absorption spectrometry. For the first time, the variability of the tropospheric part of the zenith column amount was investigated.

- High resolution spectrometers were used for lunar absorption measurement of time series during polar night.

The following knowledge on processes in the atmosphere was gained by application of retrieval theory on experiments mentioned above:

- The important role of chlorine nitrate as a reservoir for reactive chlorine and nitrogen compounds in the winter arctic stratosphere was quantified for the first time. Furthermore knowledge was gained on the temporal development during polar winter of the most important chlorine reservoirs.

- Subsidence of the arctic polar vortex was confirmed by measurement of long-lived tracers. Downwelling of air masses causes low volume mixing ratios of organic chlorine compounds in the lower stratosphere, along with increased reactive chlorine and chlorine reservoirs.

- Evidence for sequestering of nitric acid in particles or droplets in polar stratospheric clouds as well as denitrification and dehydration was found.

- It was shown that measured concentrations of dinitrogen pentoxide in the nighttime stratosphere are consistent with our knowledge on atmospheric nitrogen chemistry.

- HOCl in the nighttime stratosphere was measured, and consistence of measurements with model calculations were shown to be consistent with in the measurement error margin.
- The formation of the ClO dimer during night was proven indirectly by measurements of low amounts of molecular ClO. Consistence with model calculations was shown within the error bars.

After these sucessful infrared spectroscopy remote sensing experiments, the measurement principle is also used by the MIPAS–ENVISAT space-borne experiment of the *European Space Agency* (ESA). This is the first high spectral resolution *Michelson* interferometer ever which records limb infrared emission spectra from a space-borne platform. A dedicated analysis strategy for this experiment is presented and discussed with a focus on comparison to alternative analysis strategies. The main features of our concept are the representation of retrieval parameters on an altitude grid which is independent from the tangent altitudes of the measurement, moderate application of regularization, and an optimized selection of measurement gridpoints for analysis. These strategic considerations led to a data processor which supports numerous special applications of scientific relevance which are necessary for the analysis of much more than 30 trace species, in particular calculations for non-local thermodynamic equilibrium conditions, inclusion of line coupling, modeling of aerosol radiative transfer, as well as various options of regularization. In a blind test on the basis of synthetic measurements our data processor proved to be reliable. Due to its flexible specification it is not only applicable to MIPAS data but also supports application to data from spectroscopic experiments of different measurement geometry and spectral resolution.