# Progress in Tropospheric Ammonia ($NH_3$) Retrieval from the MIPAS Satellite Instrument

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**Abstract**

The Michelson Interferometer for Passive Atmospheric Sounding (MIPAS) is a polar orbiting high resolution mid-infrared emission limb sounder with a nominal vertical resolution of 3 km. Work to extend the list of routinely retrieved species led to the examination of ammonia, $NH_3$. In this paper, we explore the feasibility of upper tropospheric retrievals around 6 km altitude, discussing appropriate methods. We demonstrate sensitivity to latitudinal variations, using coaddition to improve signal-to-noise and compare these first results with a model.

*Key words:* MIPAS, Envisat, Ammonia, NH3, VMR, Retrieval, Troposphere

## 1 Introduction

MIPAS was launched as part of Europe's environmental monitoring satellite ENVISAT, aboard an Ariane-5 rocket on 1st March 2002. ENVISAT was injected into a polar orbit at an altitude of about 800 km and has an orbital period of about 100 minutes. MIPAS is an actively cooled atmospheric thermal emission limb sounder, working in the mid-infrared with a field-of-view that is approximately $3 \times 30$ km. MIPAS obtains high resolution spectra (0.025 $cm^{-1}$) that cover the range 685–2410 $cm^{-1}$ (14.6–4.15 $\mu$m). A complete limb scan sequence consists of 17 spectra with tangent points at 68 km, 60 km, 52 km, 47 km, 42 km and downward to 6 km in 3 km steps (ESA, 2000).

Ammonia ($NH_3$) is a simple pyramidal molecule present in small amounts in the lower atmosphere. It has two main absorbtion features in the thermal infrared, 700–1200 $cm^{-1}$ and 1500–1800 $cm^{-1}$, that can be resolved into lines. Continental mixing ratios in the lower troposphere typically range from 0.1 to 10 ppbv (Seinfeld and Pandis, 1997), repidly decreasing with altitude. The few measurements above 6 km seem to

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indicate mid-latitude VMRs of 20-50 pptv. Ammonia is the most significant basic atmospheric gas, and plays a key role in the formation and composition of aerosols through a variety of processes, many of which are poorly understood. Major natural sources include oceans, animal respiration and soil microbial processes. Significant anthropogenic ammonia emission originates from livestock, chemical fertilizers, waste management and industry. For each of these sources there remain large uncertainties in the magnitude of emissions, the diurnal and seasonal variation, and the spatial distribution. As a consequence, there are large uncertainties in the formation of sulphate and nitrate aerosols. Application of satellite data, with its excellent global coverage, would greatly enhance our knowledge of both the distribution and magnitude of these sources and sinks.

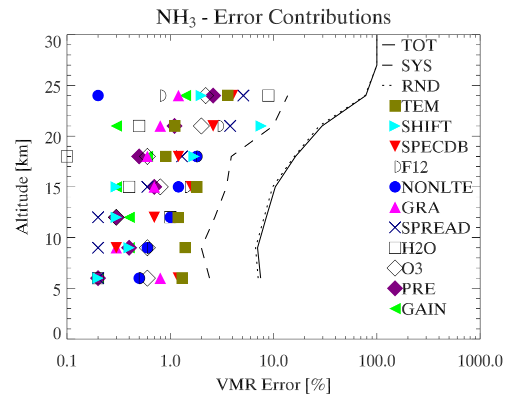


Fig. 1. Assignment of error sources to the final error budget as calculated during microwindow selection. The total error (solid line), the sum of the random error component (dotted line, based on the instrument NESR) and the total of the systematic error contributions (dashed line) are shown. Symbols show individual systematic errors, including: 'TEM' – retrieved temperature uncertainty, 'SPECDB' – uncertainty in spectroscopic line parameters, 'SPREAD' – uncertainty in the ILS & 'H2O' – water vapour uncertainty.

## 2 Retrieval Methodology

A reference climatology profile with neither seasonal nor latitudinal variation, that forms a part of the operational MIPAS processing dataset, and the Oxford Reference Foward Model (Dudhia, 2002) were used to simulate the contribution from ammonia with respect to the total expected radiance at the satellite. This radiance contribution was above the Noise Equivalent Spectral Radiance (NESR) of the instrument for many lines in the region 850 to 1250 $cm^{-1}$.

Microwindow selection enables a detailed consideration of the feasibility of retrieval. Microwindows are small regions of the spectrum, no more than 3 $cm^{-}1$ wide. Historically these have been hand-selected, but in our approach these regions are optimised to maximise the information obtained on the target species, whilst minimising both random and systematic errors. An informative discussion of the development of the microwindow selection algorithm may be found in Dudhia et al. (2002). A detailed error analysis is also obtained, showing the relative significance of all the known error sources. This analysis is shown in Figure 1, from which we conclude that, based on the climatological profile, the range over

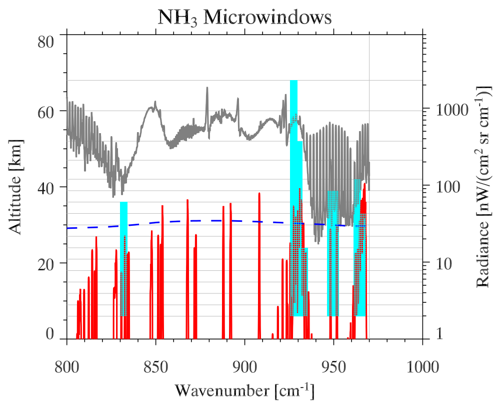


Fig. 2. The spectral positions and altitude ranges (shaded blocks, left axis) of the selected microwindows, overplotted on the total simulated radiance at 12 km tangent height (dark grey, right axis). The $NH_3$ contribution to this radiance (vertical lines) and instrument NESR (dashed line) are also shown. The MIPAS nominal measurement altitudes are shown as pale horizontal lines.

which an ammonia retrieval should be worthwhile is 6–21 km. By considering the climatological uncertainty and the total retrieval error, we may calculate the net gain in 'information' from performing a retrieval as 20.2 bits. One bit of information is defined as a halving in the uncertainty of the species concentration at one altitude level (Rodgers, 2000). Figure 2 shows these microwindows overlaid on the resultant simulated radiance for a 12 km tangent height.

The microwindow selection was validated using controlled retrievals from 'blind test' spectra, originally created as part of the E.U. Framework V 'AMIL2DA' study (AMIL2DA, 2003). Reasonable but 'unknown' profile values for all atmospheric constituents are used to model the atmospheric radiance incident at the satellite. Appropriate random noise is added and a full retrieval on this synthetic data is then performed. The resulting retrieved profile can be compared with the blind test model input to assess the accuracy of the retrieval scheme. Additionally, it enables a good check of error estimates and their influence on the retrieval. The results of this test, together with the estimated information gain implied ammonia would be a good retreival candidate.

### *2.1 Initial Retrievals*

A problem in limb profile retrievals is contamination of the lower views with cloud. If a set of retrievals is performed over an entire orbit, it becomes obvious where significant cloud is located because poor retrieval convergence is observed. There is a strong correlation between these measurements and high continuum in the spectra. One must discard contaminated spectra by choosing the point at which the damaging influence of cloud contamination on the retrieval is minimised, whilst maximising the number of low-altitude profile points retrieved. The method of cloud detection applied in this paper relies on the ratio between the radiance of two small regions of the spectrum, 788.20–796.25 $cm^{-1}$ and 832.30–834.40 $cm^{-1}$, with a threshold value of 1.8 for this ratio (Spang et al., 2004). All ratios falling below this value are flagged as cloudy. The conservative threshold of 1.8 does not take into account the continuous gradient of cloud optical thicknesses, and high cirrus may escape detection.

The retrieval of profiles from MIPAS data has been achieved using the Oxford Processor To Invert MIPAS Observations (OPTIMO) which uses the optimal estimation method (Rodgers, 2000) to find the most probable solution consistent with both the measurements and prior (a priori) knowledge. Pressure-temperature then water vapour were individually retrieved; all other interfering species were set to the climatological values used in the operational ESA MIPAS processor (Ridolfi et al., 2000).

Despite a positive feasibility assessment, the retrieval gave poor results from MIPAS data. An over-estimate in the upper troposphere of the climatological abundance used to initalise the retrieval was a possible cause, if the true ammonia VMR was falling away faster with altitude than had been anticipated. The climatological fields had used a constant tropospheric value based on boundary layer values up to the tropopause. A new a priori was created that was lower in the troposphere. A differing structure was also added, consisting of a steep falloff throughout the troposphere. It is shown in Figure 3. The lower a priori does not significantly affect the validity of the selected microwindows, but the reduced radiance impacts the random error and available information. Repeating the retrieval, with the improved a priori, showed limited improvement. The retrieval diagnostics highlighted that little information was gained. There were only 1-2 degrees of freedom per profile (defined as the trace of the averaging kernel matrix). This indicates that the useful retrieval of individual, independent profiles of ammonia are not feasible as the uncertainty on the measurement is of similar magnitude to the uncertainty in the climatology. Figure 4 shows that only the 6 and 9 km levels show significant contributions to the retrieved values. Above 12 km there is almost no sensitivity to the atmospheric state, indicating that values above this altitude are based on the a priori constraints on the retrieval. It was decided to interpolate the 6 and 9 km levels onto a constant pressure surface of 500 mb – around 6 km.

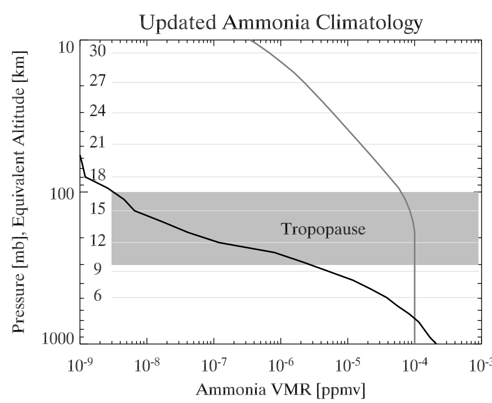


Fig. 3. The new ammonia climatology (black line) compared with the original version (grey line). Only the lower atmosphere, to 30 km, is shown. There is neither latitudinal nor seasonal variation in either climatology. Below 6 km the profile has been continued to the surface, matching typical boundary layer values that range from 0.1–1 ppbv. Ammonia is not well mixed (uniform tropospheric VMR) due to its very short lifetime.

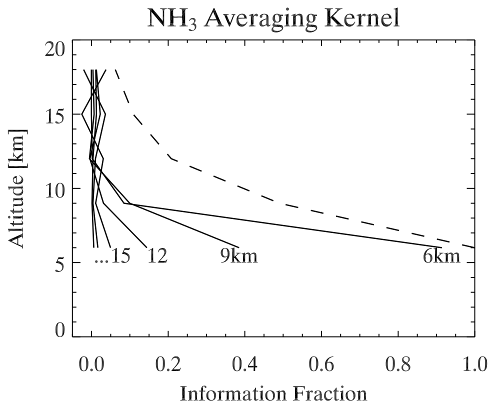


Fig. 4. Averaging kernels from a successful retrieval. The solid lines show the contribution a measurement at one altitude has made to the retrieved values at all levels. The dotted line indicates the sum of these contributions for each level and gives an indication of the total contribution from all measurements to the final retrieved value.

### *2.2 Kalman Filter*

A more complex retrieval was applied making use of a Kalman filter (Rodgers, 2000). In this approach, the final state (VMR profile, covariance etc.) of the first sucessful retrieval is used as the initial state of the subsequent retrieval. The resulting state from this second retrieval is used as the inital state for the next. This approach assumes that the variation between scans around an orbit is small. The state is 'relaxed' between profiles, so as to not over constrain the subsequent retrieval. A value that corresponds to an along-track length of 20,000 km was chosen as the relaxation factor, so measurements of one pole are (approximately) uncorrelated with those of the other pole. The amount of information in a given spectrum is unchanged, but by starting the retrieval much closer to the 'true' state the a priori influence is reduced.

## 3 Measurement & Validation

The accurate quantification of atmospheric ammonia is particularly challenging. Ammonia is a 'sticky' gas that readily adsorbs onto most surfaces, easily desorbing if ammonia levels decrease in the sample air stream. Consequently, in situ measurements are difficult to make and suffer from both under and over estimates. Remote sensing is equally difficult as the high reactivity of ammonia leads to very low VMRs in the free troposphere. Consequently, very little height-resolved information exists. The majority of prior ammonia measurements are concerned with the boundary layer and often consist of point measurements near to areas of local interest (e.g. farm emissions). The most useful form of validation was therefore a model comparison. The model itself is a global chemical model (HadAM3-STOCHEM), driven by historic advection data, (Stevenson et al., 2003). The ammonia values originate from a selection of surface point measurements used as constant source terms to initialise the model, from the 'EDGAR' database. The model contains 10 psuedo-pressure levels that reach from the surface up to the tropopause. The model resulted in 120 monthly mean global data fields, driven by 1990s dynamics. For validation purposes, a decadal mean was taken for each month to try to capture monthly and seasonal variations in the ammonia field in the troposphere.

The comparison of the ammonia field for August 2003 with a decadal mean model average for the month of August is shown in Figure 5. The model appears to have a greater dynamic range – it contains values that are up to an order of magnitude higher than the retrieval. On taking the zonal mean, the dominant difference is clearly the sucessful capturing of larger ammonia values in the equatorial regions.

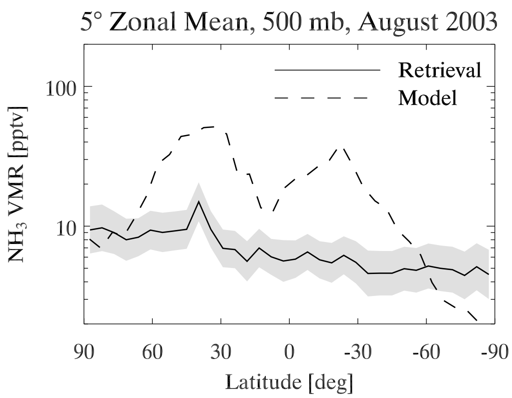


Fig. 5. Comparison of latitudinal structure of the model and retrieval for August 2003. Shading indicates the RMS of the random error.

## 4 Conclusions & Further Work

We have tried to give an impression of both the significance and difficulty of ammonia retrievals, along with a demostration of the current progress that has been made. Despite the initial setbacks, progress has been made in the retrieval of ammonia from MIPAS in the mid-troposphere. It is unusual to retrieve significantly below the a priori, with good convergence (low $\chi^2$) without some underlying physical reason. Future work would involve the retrieval and validation against model results for other months from 2003 and investigation into the lack of dynamic range of the retrieval. Finally, it is expected that the improved global ammonia distribution will aid the understanding of aerosol formation and the interaction of ammonia in sulphur and nitrogen cycles.

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