Chapter 6

Summary and Outlook

6.1 Summary

The research described in this thesis concerns satellite remote sensing of cloud properties needed to accurately retrieve information about tropospheric trace gases. Two satellite instruments developed to retrieve information about atmospheric trace gases, such as ozone and nitrogen dioxide, are the Scanning Imaging Absorption Spectrometer for Atmospheric Charactography (SCIAMACHY) and the Global Ozone Monitoring Experiment (GOME and GOME-2). Clouds have a large impact on satellite observations and therewith on trace gas retrieval results. To accurately account for clouds in trace gas retrievals, information about the relative amount of clouds within the instrument footprint (the cloud fraction), the cloud optical thickness and the cloud top pressure is needed. In this thesis, a new method to retrieve these three cloud parameters from GOME and SCIAMACHY measurements is developed. The retrieved cloud parameters are shown to improve the retrievals of ozone profiles in the troposphere. Here, the content and main conclusions of the chapters of this thesis are summarized.

In chapter 2, first the quality of the measurements of GOME and SCIAMACHY in the oxygen A-band at 760 nm, which are primarily used for the retrieval of cloud parameters, is evaluated. This is done by means of the retrieval of surface pressures from these measurements under cloud-free conditions. These retrievals can be well validated because surface pressure is a quantity that is, in general, accurately known. Therefore, surface pressure retrievals and their validation provide important insight into the quality of the instrument calibration and the forward model. In the surface
pressure retrieval we neglect aerosols. Using simulated measurements, it is shown that above low to moderate surface albedos this generally leads to an underestimation of the surface pressures, which ranges from about 50–300 hPa for surface albedos around 0.05 to about 10–50 hPa for surface albedos around 0.3. This underestimation is caused by the reduction of the light path due to scattering by aerosols. For high surface albedos (> 0.4), (multiple) scattering by aerosols can lead to an enhancement of the light path, which in turn leads to an overestimation of the surface pressure of up to 30 hPa. The magnitude of these effects is shown to be further dependent on aerosol optical thickness, the aerosol height distribution and observation geometry. Furthermore, aerosols cause a characteristic spectral feature in the fitting residuals. The amplitudes of these spectral features depend again on the aerosol optical thickness and height distribution. Surface pressures retrieved from SCIAMACHY measurements compare well with those from the UKMO meteorological dataset, taking into account the expected dependence on aerosols. However, a systematic positive offset of about 20 hPa is observed in the retrieved surface pressures. A similar offset was found in the comparison of surface pressures retrieved from SCIAMACHY and co-located GOME measurements. This bias was therefore interpreted as being caused by a calibration error on the SCIAMACHY measurements. Adding an offset of 0.86% of the continuum reflectance at 756 nm to the SCIAMACHY reflectances in the oxygen A-band removes this bias.

In chapter 3, a model is introduced to efficiently calculate the radiative transfer of polarized light in vertically inhomogeneous atmospheres that contain homogeneous cloud layers. Such calculations are generally very time-consuming. A novel approach to the radiative transfer problem in such atmospheres is taken by combining the Gauss-Seidel method, which is efficient for inhomogeneous cloudless atmospheres, with the doubling method, which is efficient for homogeneous cloud layers. For an atmosphere containing one optically thick cloud layer, this combined doubling and Gauss-Seidel (CODAGS) model is about 3–5 times faster than the doubling-adding model with an accuracy of 0.01%. The efficiency of the calculations is further increased by reducing the number of Gaussian-quadrature streams for the optically thin atmospheric layers in comparison to the number of streams used for the cloud layers. To optimize the model for radiative transfer calculations in absorption bands, the cloud reflec-
tion and transmission matrices are interpolated over the absorption and scattering optical thickness values within the cloud layers. The number of interpolation points that is needed to obtain a certain accuracy depends on the ranges of absorption and scattering optical thickness present in the evaluated wavelength range. For this, a scheme is developed to automatically determine the number of interpolation points and their distribution over optical thickness. The accuracy and efficiency of the CODAGS model, including the reduction of streams and the interpolation technique, is evaluated by means of radiative transfer calculations in the oxygen A-band and the Hartley-Huggins ozone band (295–335 nm). It is shown that SCIAMACHY measurements in these absorption bands for an atmosphere containing one optically thick cloud layer can be simulated with an accuracy better than 0.1% within 12–60 seconds on a Pentium 4 2.8 GHz PC. Overall a reduction of 2–3 orders of magnitude in the computational effort is achieved. This radiative transfer model is used for the research described in chapter 4 and 5.

In chapter 4, a new method is presented to retrieve information about cloud fraction, cloud optical thickness and cloud top pressure from GOME and SCIAMACHY measurements. The method uses measurements at the oxygen A-band in combination with measurements in the UV from 350–390 nm. It is shown that measurements of the oxygen A-band alone do not contain sufficient information to independently retrieve cloud fraction, cloud optical thickness and cloud top pressure. Adding the UV measurements allows to retrieve significant information about all three cloud parameters. Additionally, information about the surface reflection can be obtained from these measurements. Measurements in the UV add information on the cloud fraction because in this wavelength range, the spectrum of the clear-sky part of the measurement is dominated by Rayleigh scattering, which has a strong wavelength dependence. Due to the significant differences between the spectral signature of Rayleigh scattering and that of scattering by cloud particles, the UV window is especially sensitive to the cloud fraction. To illustrate the relevance of this cloud retrieval approach, it is shown that the common use of effective cloud fractions and cloud top pressures leads to a significant systematic underestimation of the tropospheric NO$_2$ Air Mass Factor in cloudy atmospheres. This underestimation can be avoided when all cloud parameters are retrieved independently, as is done with the here
developed method. The cloud retrieval is applied to GOME measurements. The results of the algorithm compare well to cloud parameters obtained by the GRAPE algorithm from co-located ATSR-2 measurements. The ATSR-2 measurements have a high spatial resolution ($1 \times 1 \text{ km}^2$) and are therefore better suited to retrieve cloud parameters, particularly the cloud fraction, from. The distributions of the differences between the cloud fractions, cloud optical thicknesses and cloud top pressures retrieved by our algorithm and the corresponding ATSR-2 values have median values and 68% confidence levels (between brackets) of -0.01 ($\pm 0.11$), 2.5 ($\pm 7.5$) and -33 ($\pm 111$) hPa, respectively.

In chapter 5, the use of the cloud parameters retrieved by the algorithm described in chapter 4 to account for clouds in ozone profile retrievals is evaluated. This approach, dubbed the CUVO\textsubscript{2} approach, is compared with two commonly used approaches in ozone retrievals, namely (1) to treat clouds as an effective ground surface albedo, commonly known as the cloud as albedo (CaA) approach; and (2) using effective cloud fractions and cloud top pressures retrieved from oxygen A band measurements assuming a cloud optical thickness of 40 (Ceff approach). By means of simulated GOME retrievals for an ensemble of realistic cloudy scenes, we show that the mean ozone concentrations in the troposphere are retrieved with errors below 3% when using the CUVO\textsubscript{2} approach, while they are generally underestimated in the tropospheric layers up to 85% when using the CaA approach and overestimated by up to 18% with the Ceff approach. Generally, the highest errors are obtained near the surface. The underestimation due to the CaA approach is caused by the neglect of the fractional cloud cover and the elevation of clouds. The Ceff approach causes an overestimation because the effective cloud fraction is a wavelength dependent quantity, as was shown in chapter 4. Then, ozone profiles are retrieved from real GOME measurements using the CUVO\textsubscript{2}, CaA and Ceff approaches and the results are validated with co-located ozonesonde measurements. The mean differences between ozone concentrations in the troposphere measured by the sondes and those retrieved using the CUVO\textsubscript{2}, CaA and Ceff approaches are found to be 15%, -60% and 27%, respectively. The unexpected overestimation of the ozone concentration when using the CUVO\textsubscript{2} approach is shown to be caused by about a third of the retrievals for which too high cloud optical thickness values ($\gg 40$) are retrieved erroneously. For these
cases, a large overestimation of the mean tropospheric ozone of up to 45% is obtained when using the CUVO$\textsuperscript{2}$ approach. The Ceff approach leads to an even larger overestimation of the ozone concentration of up to 60% for these cases. As shown in chapter 4, the retrieval of such high optical thickness values indicates horizontally inhomogeneous or multi-layered clouds to be present in the GOME footprint. Thus, the Ceff and CUVO$\textsuperscript{2}$ approaches are not adequate in case of inhomogeneous clouds. For the cases with cloud optical thickness values between 5 and 40, the CUVO$\textsuperscript{2}$ approach results in an overestimation of the mean ozone concentrations of less than 7%, while the Ceff approach leads to an overestimation of up to 25%, similarly to what is observed in the simulations. Also for cloud optical thicknesses lower than 5, the CUVO$\textsuperscript{2}$ approach results in a low mean error below ±4%. The Ceff approach, however, leads to a small but unexpected underestimation in the mean ozone concentrations for these optically thin clouds. This is possibly due to the large sensitivity of the effective cloud parameters to biases in the applied surface albedo for these optical thin clouds. For all approaches, large standard deviations of up to about 65% are found, which are probably (in part) due to the variability of the ozone concentrations within the large (960 × 80 km$^2$) GOME footprints.

The main conclusions of this thesis are:

1. The retrieval of surface pressures can be used to evaluate the calibration of the SCIAMACHY and GOME measurements in the oxygen A-band. Comparing the surface pressures retrieved from the SCIAMACHY measurements to those from the UKMO meteorological dataset, a systematic positive offset of about 20 hPa is observed in the retrieved surface pressures. A similar offset is found in the comparison of surface pressures retrieved from SCIAMACHY and co-located GOME measurements. Adding an offset of 0.86% of the continuum reflectance at 756 nm to the SCIAMACHY reflectances in the oxygen A-band removes this bias.

2. The novel approach of combining the Gauss-Seidel and doubling radiative transfer calculation methods yields a model that efficiently and accurately calculates the radiative transfer in inhomogeneous atmospheres that contain cloud layers. For radiative transfer calculations in absorption
bands, the efficiency of this model is further increased by interpolating the reflection and transmission matrices of the cloud layers over the absorption and scattering optical thickness within the cloud layers. Overall, a reduction of 2–3 orders of magnitude in the computational effort is achieved while maintaining the required accuracy ($< 0.1\%$).

3. Independent information about cloud fraction, cloud optical thickness and cloud top pressure can be retrieved by combining measurements in the oxygen A-band and in the UV between 350 and 390 nm.

4. By retrieving an effective surface albedo to take clouds into account in ozone profile retrievals, the tropospheric ozone concentrations are underestimated up to about 70% in the mean. Using effective cloud parameters in ozone profile retrievals generally leads to an overestimation of the tropospheric ozone concentrations of up to about 20% in the mean. By using the cloud parameters retrieved with the algorithm presented in this thesis, the mean differences between the tropospheric ozone concentrations retrieved from GOME measurements and those obtained from ozonesonde measurements are within 7% for most conditions.

### 6.2 Outlook

The presented method to retrieve cloud properties from measurements in the UV and the oxygen A-band was shown to be able to significantly improve tropospheric trace gas retrievals in cloudy atmospheres. The current version of the retrieval algorithm was developed to prove the feasibility of this method but is too slow to process a large amount of GOME and SCIAMACHY data. Even though quite some effort was already spent to develop an efficient radiative transfer model (chapter 3), the most time-consuming part of these retrievals is still the radiative transfer calculation. The bottleneck of these calculations is the computation of the cloud reflection and transmission matrices with the doubling method. To further reduce the computational effort of this model, the reflection and transmission matrices could be obtained efficiently from a pre-calculated look-up-table. Such a look-up-table should contain cloud reflection and transmission matrices for a variety of absorption and scattering optical thickness values. The
interpolation between these values can then be performed fast and accurately as described in chapter 3. The resulting model is then expected to be able to perform accurate radiative transfer calculations in absorption bands within about 1–10 seconds on a 2.8 GHz PC. With such an improved radiative transfer model, the cloud retrieval algorithm developed in this thesis can be expected to be fast enough to process large amounts of GOME and SCIAMACHY data.

As shown in chapter 4, the presented cloud retrieval algorithm retrieves too high cloud optical thickness values and too low cloud fractions in about 15% of the GOME measurements over land which contain inhomogeneous clouds. Furthermore, in chapter 5 large errors were obtained in the retrieved tropospheric ozone concentrations for exactly these cases regardless of the approach to account for clouds. The frequency of the problems due to horizontal cloud inhomogeneity is expected to increase with increasing footprint size. This is confirmed by the fact that the occurrence of these erroneous cloud retrievals increased by about 20% when the cloud retrievals were performed on the larger sized GOME footprints used for the ozone profile retrievals. SCIAMACHY and GOME-2 have considerably smaller footprint sizes than GOME and thus the problems due to the cloud horizontal inhomogeneity are expected to be substantially reduced when the presented cloud retrieval algorithm is applied to their measurements. Moreover, the inevitable increase of spatial resolution for future instruments will reduce the problem due to horizontally inhomogeneous clouds even further. Problems due to multi-layered clouds, however, will remain.

The sensitivity study on simulated measurements presented in chapter 4 revealed the potential of the presented cloud retrieval algorithm to retrieve cloud information above snow covered scenes. Moreover, the snow albedo can be retrieved simultaneously. Retrieval of cloud parameters above snow is of great importance since most available algorithms for GOME and SCIAMACHY are not able to distinguish clouds from snow covered surfaces. However, in the validation of the retrieved cloud parameters with ATSR-2 measurements presented in chapter 4, scenes with snow covered surfaces are excluded since the ATSR-2 retrievals are unreliable for these cases. Therefore, the quality of the retrieved cloud parameters above snow covered surfaces has not been evaluated in this thesis. In order to do so,
cloud parameters retrieved from SCIAMACHY above snow covered surfaces could be validated using MODIS, which is able to retrieve cloud optical thickness and cloud height above snow covered surfaces (King et al., 1997), although there is a time lag of about 30 minutes between the two observations. In addition, trace gas retrievals over snow covered surfaces using clouds retrieved with the presented algorithm should be validated.

The ozone profile retrievals presented in chapter 5 showed good agreement with the ozonesonde measurements in the mean. However, a large standard deviation of up to 65% was found in the troposphere, which are likely -at least partly- due to variation of ozone concentrations within the 960 × 80 km² sized GOME footprint. Ozone profiles retrieved from higher spatial resolution measurements are thus expected to show an improved comparison with ozonesondes. Moreover, as demonstrated by Sparling et al. (2006), the representation error in comparisons of ozonesondes to satellite measurements at spatial resolutions higher than 50 × 50 km² is within 10%. This in turn would enable effects of e.g. clouds on ozone profile retrievals to be studied in more detail. More importantly, well validated individual ozone profile retrievals are very valuable for studies of the physical and chemical processes in the atmosphere.

To also improve the vertical resolution of the ozone profiles retrieved from nadir measurements, the spectral fitting residuals have to be reduced. As discussed in chapter 5, the Root-Mean-Squared (RMS) values of the obtained residuals are in the order of 0.8% while the RMS of the measurement noise is 0.3%. These large residuals are probably due to errors in the forward model due to insufficient correction of the Ring effect and the ‘undersampling’ effect (van Deelen et al., 2007), errors in the ozone cross sections (Orphal, 2003) or calibration errors of the GOME measurements (van der A et al., 2002; Tanzi et al., 2002; Liu et al., 2005). Significantly more information about tropospheric ozone can be retrieved when the residuals are reduced to noise level by addressing these issues. However, then also the errors due to simplified treatments of clouds in the ozone retrieval strongly increase, as shown in chapter 5. The use of the here developed method then becomes more important.

An important application of GOME ozone profile retrievals is the validation of chemistry-transport models (e.g. de Laat et al., 2007). Until now, such validations were limited to ozone in the stratosphere because of the
low accuracy of the retrievals in the troposphere. In this thesis it is shown that the information on tropospheric ozone present in the GOME measurements (about 0.5–1.5 of the total DFS) can be accurately retrieved when clouds are properly taken into account. This allows to extend the validation of chemistry-transport models to tropospheric ozone. Furthermore, these retrievals can improve assimilations of global tropospheric ozone concentrations.