Chapter 7

Summaries

7.1 Summary

The natural nitrogen cycle has been and is significantly perturbed by anthropogenic emissions of reactive nitrogen (Nr) compounds into the atmosphere, resulting from our production of energy and food. In the last century global ammonia (NH$_3$) emissions have doubled and represent nowadays more than half of total the Nr emissions. NH$_3$ is also the principal atmospheric base in the atmosphere and rapidly forms aerosols by reaction with acids. It is therefore a species of high relevance for the Earth’s environment, climate and human health (Chapter 1). As a short-lived species, NH$_3$ is highly variable in time and space, and while ground based measurements are possible, they are sparse and their spatial coverage is largely heterogeneous. Consequently, global spatial and temporal patterns of NH$_3$ emissions are poorly understood and account for the largest uncertainties in the nitrogen cycle. The aim of this work is to assess distributions and spatiotemporal variability of NH$_3$ using satellite measurements to improve our understanding of its contribution to the global nitrogen cycle and its related effects.

Recently, satellite instruments have demonstrated their abilities to measure NH$_3$ and to supplement the sparse surface measuring network by providing global total columns daily. The Infrared Atmospheric Sounding Interferometer (IASI), on board MetOp platforms, is measuring NH$_3$ at a high spatiotemporal resolution. IASI circles the Earth in a polar Sun-synchronous orbit, covering the globe twice a day with a circular pixel size of 12km diameter at nadir and with overpass times at 9:30 and 21:30 (local solar time when crossing the equator). An improved retrieval scheme based on the calculation of Hyperspectral Range Index (HRI) is detailed in Chapter 2 and compared with previous retrieval methods. This approach fully exploits the hyperspectral nature of IASI by using a broader spectral range (800-1200 cm$^{-1}$) where NH$_3$ is optically active. It allows retrieving total columns from IASI spectra globally and twice a day without large computational resources and with an improved detection limit. More specifically the retrieval procedure involves two steps: the calculation of a dimensionless spectral index (HRI) and the conversion of this index into NH$_3$ total columns using look-up tables (LUTs) built from forward radiative transfer simulations under various atmospheric conditions. The retrieval also includes an error characterization of the retrieved column, which is of utmost importance for further analysis and comparisons. Global distributions using five years of data (1 November 2007 to 31 October 2012) from IASI/MetOp-A are presented and analyzed separately for the morning and evening overpasses. The advantage of the HRI-based retrieval scheme over other methods, in particular to identify smaller emission sources and transport patterns over the oceans is shown. The benefit of the high spatial sampling and resolution of IASI is highlighted with the regional distribution over China and the first four-year time series are briefly discussed.

We evaluate four years (1 January 2008 to 31 December 2011) of IASI-NH$_3$ columns from the morning observations and of LOTOS-EUROS model simulations over Europe and Western Russia. We describe the methodology applied to account for the variable retrieval sensitivity of IASI measurements in Chapter 3. The four year mean distributions highlight three main agricultural hotspots in Europe: The Po Valley, the continental part of
CHAPTER 7. SUMMARIES

Northwestern Europe, and the Ebro Valley. A general good agreement between IASI and LOTOS-EUROS is shown, not only over source regions but also over remote areas and over seas when transport is observed. The yearly analyses reveal that, on average, the measured NH$_3$ columns are higher than the modeled ones. Large discrepancies are observed over industrial areas in Eastern Europe and Russia pointing to underestimated if not missing emissions in the underlying inventories. For the three hotspots areas, we show that the seasonality between IASI and LOTOS-EUROS matches when the sensitivity of the satellite measurements is taken into account. The best agreement is found in the Netherlands, both in magnitude and timing, most likely as the fixed emission timing pattern was determined from experimental data sets from this country. Moreover, comparisons of the daily time series indicate that although the dynamic of the model is in reasonable agreement with the measurements, the model may suffer from a possible misrepresentation of emission timing and magnitude. Overall, the distinct temporal patterns observed for the three sites underline the need for improved timing of emissions. Finally, the study of the Russian fires event of 2010 shows that NH$_3$ modeled plumes are not enough dispersed, which is confirmed with a comparison using in situ measurements.

Chapter 4 describes the comparisons of IASI-NH$_3$ measurements with several independent ground-based and airborne data sets. Even though the in situ data are sparse, we show that the yearly distributions are broadly consistent. For the monthly analyzes we use ground-based measurements in Europe, China and Africa. Overall, IASI-derived concentrations are in fair agreement but are also characterized by less variability. Statistically significant correlations are found for several sites, but low slopes and high intercepts are calculated in all cases. At least three reasons can explain this: (1) the lack of representativity of the point surface measurement for the large IASI pixel, (2) the use of a single profile shape in the retrieval scheme over land, which does therefore not account for a varying boundary layer height, (3) the impact of the averaging procedure applied to satellite measurements to obtain a consistent quantity to compare with the in situ monthly data. The use of hourly surface measurements and of airborne data sets allows assessing IASI individual observations. Much higher correlation coefficients are found in particular when comparing IASI-derived volume mixing ratio with vertically resolved measurements performed from the NOAA WP-3D airplane during CalNex campaign in 2010. The results demonstrate the need, for validation of the satellite columns, of measurements performed at various altitudes and covering a large part of the satellite footprint.

The six-year of IASI observations available at the end of this thesis are used to analyze regional time series for the first time (Chapter 5). More precisely, we use the IASI measurements over that period (1 January 2008 to 31 December 2013) to identify seasonal patterns and inter-annual variability at subcontinental scale. This is achieved by looking at global composite seasonal means and monthly time series over 12 regions around the world (Europe, Eastern Russia and Northern Asia, Australia, Mexico, South America, 2 sub-regions for Northern America and South Asia, 3 sub-regions for Africa), considering separately but simultaneously measurements from IASI morning and evening overpasses. The seasonal cycle is inferred for the majority of these regions. The relations between the NH$_3$ atmospheric abundance and emission processes is emphasized at smaller regional scale by extracting at high spatial resolution the global climatology of the month of maxima columns. In some region, the predominance of a single source appears clearly (e.g. agriculture in Europe and North America, fires in central South Africa and South America), while in others a composite of source processes on small scale is demonstrated (e.g. Northern Central Africa and Southwestern Asia).

Chapter 6 presents the achievements of this thesis, as well as ongoing activities and future perspectives.