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Measuring atmospheric ammonia with remote sensing campaign: Part 1 - Characterisation of vertical ammonia concentration profile in the centre of The Netherlands



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ABSTRACT

Ammonia (NH₃) is difficult to monitor at atmospheric concentrations due its high solubility and reactivity and the strong spatial and temporal variations of its concentrations. Monitoring is mostly performed using passive samplers or filter packs with daily coverage at best. Only at a few sites ammonia is measured with more expensive wet chemical or spectroscopic measurement techniques. Instruments using an open path show the most potential as these avoid the use of inlets and thus the interactions of NH₃ with tubing, filters, and inlets. Measurements on the vertical distribution of NH₃ are even scarcer. with only a few available airborne and tower measurements. Satellite observations of NH₃ show potential to be used for real-time monitoring as these have global coverage often with daily overpasses. Unfortunately, validation of satellite NH₃ products representing the total atmospheric column with ground based instruments measuring in situ NH₃ has been troublesome due to a lack of knowledge about the vertical distribution. Validation with FTIR instruments has shown potential but has been performed for only a limited number of stations. In this study we report on measurements performed during the Measuring atmospheric Ammonia with Remote Sensing (MARS) field campaign at Cabauw, the Netherlands. The aim of the campaign was to improve the general understanding of the vertical distribution of NH₃. An approach was taken using four mini-DOAS instruments installed in the meteorological tower at Cabauw, supplemented by measurements with a MARGA and a mobile FTIR instrument. The measurements between May and October 2014 showed large variations in the concentrations and maximum concentrations reached up to 240 μ g m⁻³. The lower three mini-DOAS and MARGA measurements showed large differences on an hourly basis, which were shown to originate from multiple measurement artefacts of the MARGA. The mini-DOAS concentrations varied sharply between the different levels with the lower three instruments showing maxima at night while the mini-DOAS at 160 m in the tower showed maxima during the day. The study shows that the daytime boundary layer is well-mixed with only small gradients between concentrations measured at various heights. Differences were found in the origin of the NH₃ with the instrument at 20 m showing higher concentrations with transport from the north, where the largest nearby sources are located, while the instrument at 160 m showed the highest concentrations coming from the east which shows a more regional influence. The measurement campaign data is freely available for model and satellite validation studies.

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1. Introduction

The unprecedented levels of reactive nitrogen in the earth system is one of the biggest threats to our society and environment (Rockstrom et al., 2009). Fowler et al. (2013) recently estimated that current (21st century) reactive nitrogen emissions to the atmosphere are up to four times higher than pre-industrial levels and result in a cascade of environmental effects (Galloway et al., 2003). Ammonia (NH₃) can be considered as the primary form of reactive nitrogen (Nr) input to the environment (Sutton et al., 2013). The availability of mineral fertilizer originating from industrial production of NH₃ is essential to feed the world population (Erisman et al., 2008). Besides agriculture, NH₃ is emitted by a large variety of secondary sources including biomass burning, industry, and cars equipped with a three-way catalyser (Galloway et al., 2003; Erisman et al., 2008). NH₃ is removed from the atmosphere through dry and wet deposition and has a limited atmospheric lifetime of hours up to a few days. NH₃ also reacts easily with nitric and sulphuric acids, forming ammonium salts in the process (Fowler et al., 2009). Both the deposition and chemistry pathways cause a number of environmental and health hazards. Deposition of NH₃ leads to soil acidification and enhances eutrophication causing species loss and ecosystem stress (Bobbink et al., 2010; Erisman et al., 2013). The ammonium salts are a major contributor to atmospheric particulate matter levels which are suspected to cause various health effects (Pope et al., 2002; Pope et al., 2009). Furthermore, aerosols in the atmosphere are an important factor in climate change affecting the global radiance budget through scattering solar radiation and influencing the albedo and life time of clouds (Adams et al., 2001; Myhre et al., 2013).

Although NH₃ is a major player in various environmental and health issues our knowledge of its global budget and distribution is still relatively poor (Sutton et al., 2013). This is caused by the complexity of the N-cascade combined with a lack of representative atmospheric measurements. Even though agriculture is by far the largest emission source, there are large uncertainties in both global and regional emission estimates (Galloway et al., 2008; Reis et al., 2009; Sutton et al., 2013). NH₃ emissions depend strongly on the agricultural practice in a given region. In addition, meteorological conditions have a strong impact on the emission variability as NH₃ volatilization is strongly affected by e.g. temperature, wind speed, humidity (Sommer et al., 1991). In addition, agricultural activities such as manure application depend on growing season parameters and soil conditions. Although dry deposition is a major sink for NH₃ near sources, ecosystems that are highly saturated with NH₃ may become daytime sources when the partial pressure of NH₃ becomes higher than the atmospheric concentration (Fowler et al., 2009). Removal of the NH₃ from the atmosphere depends on land use, surface state and precipitation levels. The chemical loss of NH₃ depends largely on pollution regime and links the fate of NH₃ to the sulphur and oxidized nitrogen cycle. Realizing that the spatial and temporal variability of NH₃ concentrations is large due to the primary nature of NH₃ and its short atmospheric lifetime, amplifies the need for a high resolution monitoring system with global coverage.

To monitor global distributions of air pollutants satellite remote sensing has shown great potential for pollutants such as aerosols and nitrogen dioxide as they provide (sub-daily) coverage (Boersma et al., 2007; Remer et al., 2005). Recent developments have enabled to retrieve NH₃ column densities from satellite instruments like the Tropospheric Emission Spectrometer (TES, Beer et al., 2008; Shephard et al., 2011), the Atmospheric Infrared Sounder (AIRS, Warner et al., 2016) the Cross-track Infrared Sounder (CrIS, Shephard and Cady-Pereira, 2015), and the Infrared Atmospheric Sounding Interferometers (IASI, Clarisse et al., 2009). Unfortunately, the validation of the emerging NH₃ products has been very limited so far. Promising results have been obtained using Fourier Transform InfraRed spectroscopy (FTIR) observations (Dammers et al., 2016), but only for a limited number of stations located far away from source areas. Validation of the satellite data with ground based data is complicated. Firstly, no global coverage is provided as almost all measurement locations of NH₃ are situated in Europe, North America and China. Secondly, measuring NH₃ reliably at ambient concentration levels is complicated due the reactive and sticky nature of the molecule which may lead to measurement artefacts (von Bobrutzki et al., 2010). Hence, current monitoring networks either exploit (passive) low cost systems with long sampling times or expensive annular wet denuder systems with hourly or daily coverage (Fowler et al., 2009). Scientific developments to provide high temporal resolution data include spectroscopic measurements using techniques such as Quantum Cascade Laser absorption systems (QCL), Differential Optical Absorption spectroscopy (DOAS) and Cavity-ring down spectroscopy (CRDS) (Ellis et al., 2011; Berden et al., 2000; Harren et al., 2000; Pogany et al., 2009). Currently, the open path techniques show most promising results to determine NH₃ as they avoid all interactions of NH₃ with inlet, tubing as well as volatilization of ammonium nitrate for example stuck on filters. Reliable high resolution data on NH₃ is only now becoming available for Switzerland and in the Netherlands, in which mini-DOAS instruments are employed in the monitoring networks. Thirdly, the mismatch in representativity of the ground based and satellite measurements further hampers the evaluation of satellite products as illustrated by Van Damme et al. (2015). They concluded that information on vertical profiles is highly needed to be able to estimate surface concentrations from column totals. Only a few studies report on the vertical profile of NH₃ from towers (Erisman et al., 1967; Li et al., 2017). In addition, data from a limited number of short term airborne campaigns are available (Nowak et al., 2007, 2010; Leen et al., 2013). In short, new experimental datasets with vertical profile information on NH₃ in agricultural source areas are required to improve the validation of remote sensing products.

The study presented here aims to improve the general understanding on the vertical distribution of NH₃, the processes driving the concentrations in the surrounding region and provide high temporally resolved measurements to help in the validation of the IASI-NH₃ satellite product. We report the results of the *Measuring* atmospheric Ammonia with Remote Sensing (MARS) field campaign at Cabauw, the Netherlands, and explore the use of the observations to characterize the vertical distribution of NH₃. This paper aims to describe the measurements and its uncertainties, characterize the site and the bias in the measurements and the inter-comparison between the measurements. For the campaign four mini-DOAS instruments, a MARGA instrument and a mobile FTIR instrument were installed near- and at various levels in-the meteorological tower at the CESAR supersite in the Netherlands. The Cabauw Experimental Site for Atmospheric Research (CESAR) site is well situated to explore the high variability of NH₃ for a high NH₃ emission region as it provides a wide range of complementary meteorological and air pollutant data. In section 2, the measurement site and the instrumental set-up for the field campaign is described. Section 3 reports on the results. Finally, section 4 covers a final discussion and conclusions.

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