Atmospheric Environment 148 (2017) 352–360

Contents lists available at ScienceDirect

Atmospheric Environment

journal homepage: www.elsevier.com/locate/atmosenv

Trends in ammonia measurements in the Netherlands over the period 1993–2014

M.C. van Zanten*, R.J. Wichink Kruit, R. Hoogerbrugge, E. Van der Swaluw, W.A.J. van Pul

National Institute for Public Health and the Environment, The Netherlands

HIGHLIGHTS

- Trends in observed ammonia and reported emissions between 1990 and 2004 are consistent.
- Trends between 2005 and 2014 diverge: emissions further decline but concentrations rise.
- Trends in ammonium in aerosol and precipitation do not match the trends in ammonia.
- No trend in ammonia exists due to a trend in meteorological conditions.

ARTICLE INFO

Article history:
Received 8 July 2016
Received in revised form
2 November 2016
Accepted 3 November 2016
Available online 5 November 2016

Keywords:
Ammonia
Ammonium aerosol
Wet deposition
Measurements
Trends
Emissions

ABSTRACT

We present measurements of atmospheric concentrations of ammonia and ammonium in the Netherlands over the period 1993–2014 and measurements of wet deposition of ammonium for 1985–2014. The various time series have been obtained at 16 monitoring stations from the Dutch National Air Quality Monitoring Network. The monitoring stations are geographically homogenously spread over the Netherlands and are equally distributed over regions with relatively low, moderate and high ammonia emission.

During the period covered, changes in the monitoring have occurred. To obtain consistent time series, data are revalidated or corrected when necessary, according to current validation procedures or latest technical insights. The time series of ammonia concentrations are gap filled and time series corrected for meteorological influences are constructed.

The course in the ammonia concentrations shows roughly two periods. For 1993–2004, the ammonia concentrations show a downward trend of 36%, which is statistically significant with a confidence interval (CI) of 99%. For 2005–2014, an upward trend of 19% (CI 90%) is reported. Correcting time series of ammonia concentrations for meteorological influences enhances the statistical reliability of the derived trends. This resulted in trends of −40% (CI 99%) and 24% (CI 95%) respectively. For the full period there exists no trend in ammonia concentrations due to a trend in atmospheric conditions. For 2005–2014 ammonia concentrations increased especially in springtime, while showing no change in winter months. After correcting for meteorological influences, all seasons in this period show an increase in ammonia concentrations although the increase in the spring months is still the largest.

For 1993–2014 the reported ammonia emissions in the Netherlands declined in both periods with respectively 51% and 22%. The trends in emissions and ammonia concentrations correspond in the period 1993–2004 whilst over the period 2005–2014, the trends in emissions and concentrations of ammonia diverge. This divergence is for roughly a third accounted for by processes related to changes in chemical climate (see accompanying modelling paper by Wichink Kruit et al., 2016) but it is not clear what explains the remaining difference in trends.

For 1993–2014, downward trends of wet deposition of ammonium and ammonium in aerosol are found to be 47% and 68%, respectively. A split into two periods is not found as is the case with the ammonia concentration. However, although statistically not significant, both wet deposition of ammonium and ammonium in aerosol show a leveling off in decline between 2005 and 2014.

© 2016 The Authors. Published by Elsevier Ltd. This is an open access article under the CC BY-NC-ND license (http://creativecommons.org/licenses/by-nc-nd/4.0/).

* Corresponding author.
E-mail address: margreet.van.zanten@rivm.nl (M.C. van Zanten).

http://dx.doi.org/10.1016/j.atmosenv.2016.11.007
1352-2310/© 2016 The Authors. Published by Elsevier Ltd. This is an open access article under the CC BY-NC-ND license (http://creativecommons.org/licenses/by-nc-nd/4.0/).

1. Introduction

Already in the 1990s, hourly ammonia concentrations (NH_3) were measured continuously at several monitoring stations in the Netherlands (Buijsman et al., 1998). This observation network was set up to monitor the effect of Dutch policy measures on the reduction of ammonia emissions. In the Netherlands, ammonia concentrations are among the highest in Europe due to a high density of agricultural activities. High NH_3 detriments vegetation such as lichens and leads to deposition in nature areas. Nitrogen deposition above a critical load leads to acidification and eutrophication causing a loss of biodiversity (e.g. Bobbink et al., 2010). In order to mitigate these high nitrogen deposition values, several abatement policies have been implemented over the years, of which the techniques to reduce ammonia emissions caused by housing livestock, and storing and applying of manure are best known.

As a result of the abatement policies, reported ammonia emissions show a steady decline since 1990 with the largest decline taking place in the first decade (Fig. 1). The decline is largely due to the measure of incorporating animal manure into the soil instead of broadband spreading. In the Netherlands, the total ammonia emission consists for about 85% of agricultural emissions, the latter being calculated with the National Emission Model for Agriculture (NEMA, Bruggen van et al., 2011; Velthof et al., 2012). Since 2011, the Dutch Pollutant Release and Transfer Register uses NEMA for its annual reporting of ammonia emissions from agriculture (Maas van der et al., 2011) to the European Commission (NEC Directive) and United Nations (Gothenburg protocol). The emission time series is updated every year according to the latest insights, in this article the time series as reported in 2014 are used.

The main question discussed in this paper is whether the reported decline in ammonia emissions is reflected in atmospheric concentration time series. The relation between emissions and concentration of NH_3 is not straightforward since emission characteristics and several atmospheric processes determine NH_3 in the atmosphere. After ammonia is emitted, it is subsequently mixed and transported in the atmosphere. It is chemically transformed into secondary inorganic aerosols such as ammonium nitrate and ammonium sulfate and it is removed from the atmosphere by dry and wet deposition. Finally, meteorological processes influence the effectiveness of the previously mentioned processes. Yet, atmospheric concentration of NH_3 is the most reliable component to monitor the emission trends over the years (Jaarsveld van et al., 2000; Sutton et al., 2003; Bleeker et al., 2009).

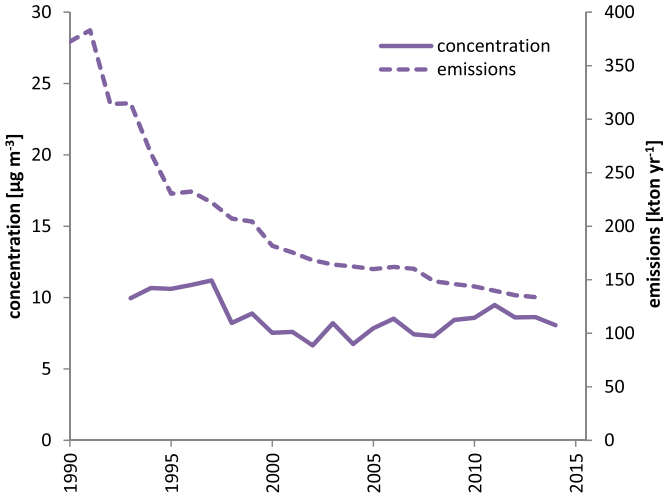

Fig. 1. Trends in reported ammonia emissions (Jimmink et al., 2015) and measured NH_3 between 1990 (emissions)/1993(measurements) and 2014. The ammonia concentration is represented by the mean of gapfilled time series of 8 monitoring stations (see Materials and methods for more information).

Because ammonium in aerosol ($(NH_4^+)_{air}$) and wet deposition of ammonium ($(NH_x)_{wet}$) suffer from additional factors making them less suitable indicators than NH_3. $(NH_4^+)_{air}$ is formed from NH_3 and as such a secondary product which is also influenced by trends in the precursors of the sulfate and nitrate parts. Especially, the rapid decline in SO_2 concentrations has an influence on the ammonium trend (see for example, Sutton et al., 2003; Horvath and Sutton, 1998). Secondly, $(NH_4^+)_{air}$ has a longer lifetime in the atmosphere than NH_3 which ensures a larger contribution of foreign emissions to its concentration in the Netherlands. Model results show 40% of $(NH_x)_{wet}$ to originate abroad (Van der Swaluw et al., 2011) while for $(NH_4^+)_{air}$ this number raises to 50%. For NH_3 this number is on average roughly 15%, though close to the German and Belgian border this number obviously may rise.

Because NH_3 is influenced by several factors as mentioned above, model calculations are necessary to account for all these processes. Wichink Kruit et al. (2016) give in an accompanying paper a detailed analysis of the trend in NH_3 using the Operational Priority Substances (OPS) model (Sauter et al., 2015), which is an atmospheric transport and deposition model. In the current article we explore what can be inferred about the reported decline in ammonia emissions from ammonia measurement analyses alone.

In this paper we will present time series of all three components NH_3, $(NH_4^+)_{air}$ and $(NH_x)_{wet}$ and derive linear trends for various periods. The data sets were partly revalidated and re-analyzed for this study. The influence of meteorological processes on NH_3 is quantified through a multivariate regression analysis of measurements of meteorological parameters. Finally, we will discuss the trends in ammonia measurements with respect to reported emission reductions.

2. Materials and methods

The Dutch National Air Quality Monitoring Network (LML, Landelijk Meetnet Luchtkwaliteit, www.lml.rivm.nl) measures various air quality components. This article discusses all measurements related to ammonia in the period 1993–2014: ammonia concentrations (NH_3), ammonium in aerosol ($(NH_4^+)_{air}$) and the wet deposition of ammonium ($(NH_x)_{wet}$). For wet deposition a time series starting in 1985 is taken. An overview of the monitoring stations and the components measured is given in Table 1 and the configuration of the monitoring stations is presented in Fig. 2.

2.1. Air concentrations of ammonia

In the network, hourly ammonia concentrations have been measured since 1993 at eight monitoring stations and since 2014 at six locations. Continuous-flow denuders, named AMORs (Amanda for MOnitoring RIVM developed by ECN, Wyers et al., 1993) are used. AMORs have a concentration range for ammonia in air of 0.5–500 $\mu g/m^3$. The uncertainty in the observed annual mean values has been estimated to be 7% (Blank, 2001). The instrument samples air by leading the airflow through a fluid in a rotating denuder tube. Ammonia is absorbed by this fluid and electrical conductivity is determined as a measure for the ammonia concentration. AMORs are stable and accurate instruments, but expensive in maintenance and use of chemicals. At the two monitoring stations where AMOR measurements have ended, triplets of Gradko passive samplers (Lolkema et al., 2015) are used for monthly mean values; in this study the measurements for the year 2014 have been included.

Download English Version:

https://daneshyari.com/en/article/5753519

Download Persian Version:

https://daneshyari.com/article/5753519

Daneshyari.com