# Effectiveness of two decades of policy measures to reduce ammonia emissions in the Netherlands

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**Abstract**
In this study, the effectiveness of two decades of policy measures to reduce ammonia emissions in the Netherlands is evaluated. It is shown that the ammonia concentration is more suitable for monitoring policy measures than the ammonium aerosol concentration or the wet deposition of $NH_x$. This study shows that the decrease in ammonia emissions due to policy measures between 1990 and present did not result in a proportional decrease of the ammonia concentration in the air. The less effectively declining ammonia concentrations can largely be explained by the change in atmospheric chemical and meteorological conditions. The large decline in oxidized sulfur and nitrogen concentrations has led to reduced formation of sulfuric and nitric acid and consequently reduced formation of ammonium salts. In this way, relatively more ammonia remained in the atmosphere. Simultaneously, the absorbing surface became less acid, which resulted in less deposition of ammonia and more ammonia remaining in the atmosphere. Meteorology has a significant effect on the year-to-year variation in ammonia concentrations, but does not significantly affect the trend in the ammonia concentrations over the years. It is likely, however, that ammonia concentrations will increase due to climate change, e.g., dryer and warmer springs/summers.

**Key Words**
Ammonia concentrations, ammonia emissions, policy measures, OPS model, CLRTAP, climate change

## Introduction

During the last 3 decades, ammonia is increasingly being recognized as an important atmospheric pollutant. This is the result of the acidifying and eutrophying effects of ammonia deposition onto terrestrial and aquatic ecosystems and the contribution of ammonia to secondary inorganic aerosol that may have detrimental effects on public health. Especially, eutrophication and acidification of soils induce shifts in the nutrient balance of plant species that can result in a loss of biodiversity. Therefore, one of the main items in the national air pollution policy of the Netherlands is to reduce agricultural ammonia emissions (as the main source of ammonia in the Netherlands). Several policy measures have been taken between 1990 and the present. The sharp decline between 1990 and 1995 (dashed purple line in Figure 1) is mainly due to reduction measures in manure application. In this period, mandatory low-emission manure spreading techniques were introduced. From 1998 on, the MINAS mineral accounting system was introduced, which reduced total ammoniacal nitrogen (TAN) content in animal manure considerably and led to an additional reduction of ammonia emissions.

To follow the effect of policy measures, $NH_3$ concentrations, $NH_4^+$ concentrations and wet deposition of $NH_x$ are measured within the National Air Quality Monitoring Network in the Netherlands from 1990 onwards. All these indicators are to a greater or lesser extent directly influenced by several processes: emissions from national and foreign sources, atmospheric transport, chemical conversion and deposition. The ammonia concentration in the air, for example, depends on other compounds that are present in the atmosphere with which it can chemically react and meteorological conditions and does consequently not have to be proportional to the ammonia emission. Figure 1 shows the development of the reported ammonia emissions (dashed purple line) and the observed $NH_3$, $SO_2$ and $NO_2$ concentrations (solid purple, red and green lines) in the Netherlands since the early nineties. Although the emissions of ammonia dropped by more than 50% since the early nineties, the observed ammonia concentrations show a much more moderate trend. In the same period, the $SO_2$ and $NO_2$ concentrations in the Netherlands have dropped by roughly 90% and 40%, respectively, as a result of the emission reductions as agreed in the Protocols under the UNECE Convention on Long-Range Transboundary Air Pollution and the EU Directive on National Emission Ceilings (NEC).

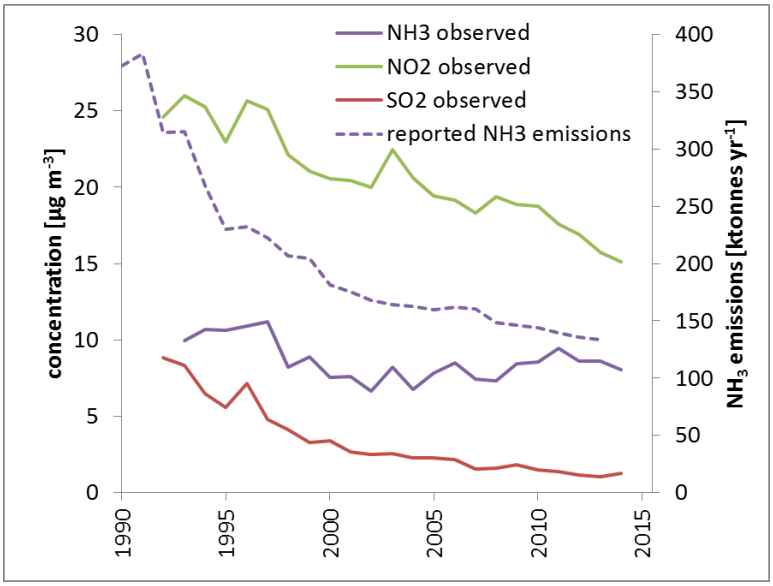


**Figure 1. Trends in reported $NH_3$ emission and observed annual average $NH_3$, $NO_2$ and $SO_2$ concentrations between 1990 and 2014.**

To draw any conclusions on the effectiveness of policy measures to reduce the ammonia emissions, the effect of the chemical and meteorological circumstances on the processes that affect the concentration in the air need to be quantified. This is done by using the Operational Priority Substances (OPS) model in which all processes are parameterized.
In this study, we will first present the general model performance of the OPS model for the available $NH_3$ concentrations, $NH_4^+$ concentrations and wet deposition of $NH_x$ in the Netherlands from 1990 till 2014. Then, we will show why $NH_3$ is the best indicator for monitoring ammonia policy measures. Finally, we will present the sensitivity of the model (and thus the observations) to changes in chemical circumstances and meteorology.

## Methods

To investigate the effects of the chemical and meteorological circumstances on the the ammonia concentration in the air, the OPS model (OPS-LT version 4.4.4) was used. The OPS model is a long-term Lagrangian transport and deposition model that describes relations between individual sources or source areas and individual receptors by Gaussian plumes. The OPS model simulates the emission, dispersion, transport, chemical conversion and deposition as a function of meteorological conditions. With OPS the annual concentration and deposition of pollutants (e.g. particulate matter, acidifying compounds like $SO_2$, $NO_x$ and $NH_3$) for the Netherlands at a high spatial resolution are calculated (typically 1 x 1 $km^2$, e.g. Velders et al., 2015; Sauter et al., 2015).
The model results of OPS-LT version 4.4.4 (red lines) are validated with observations (black lines) in the Netherlands from 1990 till 2014 for (Figure 2):

- ammonia concentrations in air, i.e., $(NH_3)_{air}$ (solid lines),
- ammonium concentrations in aerosol, i.e., $(NH_4^+)_{air}$ (dashed lines),
- wet deposition of ammonium, i.e., $(NH_x)_{wet}$ (dotted lines)

In general, the levels and the trends in $(NH_3)_{air}$ , $(NH_4^+)_{air}$ and $(NH_x)_{wet}$ are reproduced reasonably well by the OPS model. However, for $(NH_3)_{air}$, the OPS model is systematically about 1.5 - 2 µg $m^{-3}$ higher than the observations over the whole period except for the last couple of years. As a result, the trend in the model calculations (-41%) is larger than the trend in the observations (-22%) over the period between 1993 and 2014. On the other hand, the $(NH_4^+)_{air}$ is modelled typically about 0.4 µg $m^{-3}$ lower than the observations over the whole period. For this component, the trend in the modelled $(NH_4^+)_{air}$ nicely follows the trend in the observations, i.e., -86% versus -90%. Note that the decrease in $(NH_4^+)_{air}$ is much larger than the decrease in $(NH_3)_{air}$ due to the large reductions in $SO_2$ and $NO_x$ emissions and therefore reduced particle formation. $(NH_x)_{wet}$ shows a mixed picture with an overestimation by the model in the early 90s, an underestimation around the 00s and a good model performance in recent years. The trends in the modelled and observed $(NH_x)_{wet}$ differ about 10%, i.e., 55% versus 45% decrease.

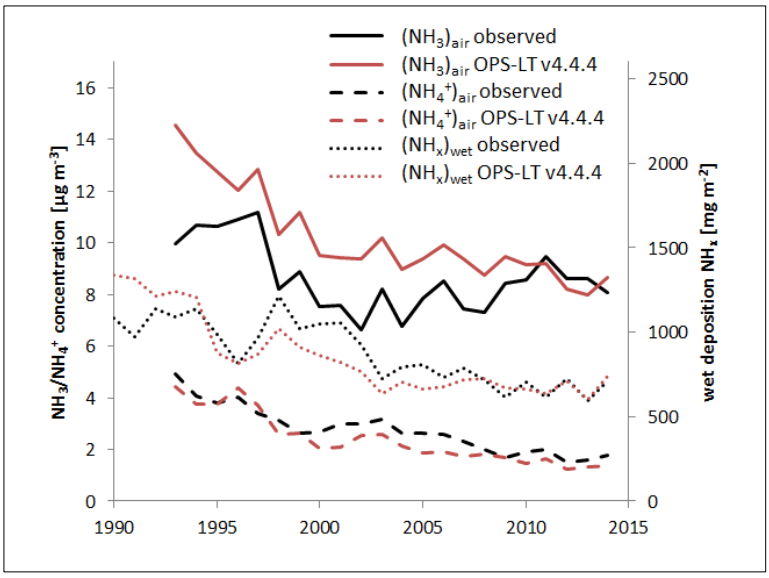


**Figure 2. Annual average at measurement stations of $(NH_3)_{air}$ (in 1993: n=5, from 1994-1999: n=7, from 2000: n=8, upper panels), $(NH_4^+)_{air}$ (in 1993: n=6, from 1994-2012: 7, from 2013: n=4; middle panels) and $(NH_x)_{wet}$ (until 2012: n=10, in 1993 and 1994: N=9, from 2013: n=6; lower panels).**

## Results

*NH3 as best indicator*

The OPS model has also been used to investigate which of the three components is the best indicator for monitoring policy measures taken in the Netherlands. For this purpose, the modelled contributions of the sources within the Netherlands to the total concentration or wet deposition were calculated. Figure 3 shows that the ammonia sources in the Netherlands contribute to about 87% of the concentration that is observed at the 8 national air quality monitoring stations at which $NH_3$ is measured; for $NH_4^+$ this is about 55%, while for the wet deposition of $NH_x$ about 60% is originating from within the Netherlands. The figure also shows that although the contribution of the sources within the Netherlands is steadily decreasing from 95% in 1990 to 87% in 2014, $NH_3$ is the best indicator to follow the policy measures taken in the Netherlands. It should be noted though that the validity of using ammonia concentrations to assess policy measures depends on the availability of measurements at a sufficient number of locations for the average concentration to reflect the average conditions.

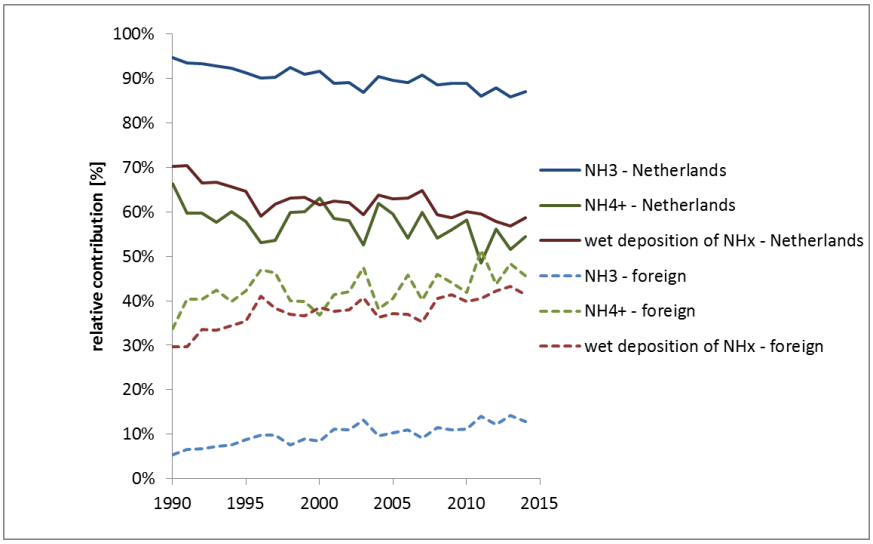


**Figure 3. Relative contribution of $NH_3$ emissions from the Netherlands and abroad to the indicators $NH_3$ concentration, $NH_4^+$ concentration and wet deposition of $NH_x$.**

*Sensitivity of the ammonia concentrations for chemical circumstances and meteorology*

Finally, the sensitivity of the observed $NH_3$ concentration to changes in chemical circumstances and meteorology was investigated by carrying out several sensitivity runs with the OPS model. The effects of the sensitivities were superimposed on the original observations to investigate how the concentrations would have looked like, when these effects would have been constant in time or would not have been occurred.

The three effects that were investigated in this study were:

- chemical conversion from $NH_3$ to $NH_4^+$ (chemical conversion rates of 2012 were fixed for all years),
- co-deposition of $NH_3$ with $SO_2$ (higher $SO_2$ levels in the past will lead to a more acid surfaces on which $NH_3$ is easier deposited) and
- meteorology (long-term meteorology, i.e., the average over 1995-2004, averages out extreme years)

To illustrate how the observed concentrations would have looked like when there would have been no change in the chemistry, no codeposition of $NH_3$ and $SO_2$ and no interannual variation in meteorology, the effects of these processes have been superimposed to the observed concentrations in the Netherlands. The resulting annual average virtual $NH_3$ concentration is shown as a dotted line in Figure 4. The reported $NH_3$ emission and the original observed $NH_3$ concentrations are shown with dashed and solid lines, respectively. The figure clearly shows that the trends in the reported emissions and the virtual $NH_3$ concentrations between 1993 and 2004 are in good correspondence, except for the first two years in which the trend in the emissions is clearly steeper. From 2005 onwards, the trends in the emissions and the concentrations have an opposite direction.

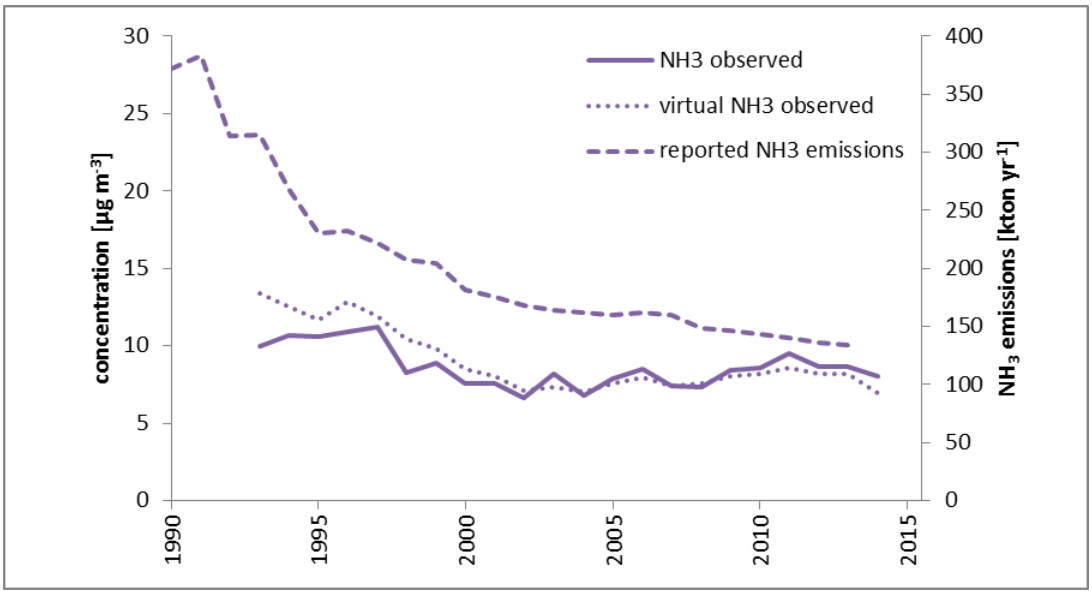


**Figure 4. Trends in reported $NH_3$ emission (dashed line) and observed annual average $NH_3$ concentration (solid line) between 1990 and 2014. The dotted line represents the annual average virtual concentration when there would not have been a change in the chemistry, codeposition of $NH_3$ and $SO_2$ and interannual variation in meteorology.**

## Conclusion

The ammonia concentration is more suitable for monitoring policy measures than the ammonium aerosol concentration or the wet deposition of $NH_x$. The observed ammonia concentrations are however not proportional to the ammonia emissions due to change in atmospheric chemical and meteorological conditions. Therefore, policy measures on ammonia emission reduction between 1990 and present did not result in a proportional decrease of the ammonia concentration in the air.

The large decline in oxidized sulfur and nitrogen concentrations has led to reduced formation of sulfuric and nitric acid and consequently reduced formation of ammonium salts. In this way, relatively more ammonia remained in the atmosphere. Simultaneously, the absorbing surface became less acid, which resulted in less deposition of ammonia and more ammonia remaining in the atmosphere. Meteorology has a significant effect on the year-to-year variation in ammonia concentrations, but does not significantly affect the trend in the ammonia concentrations over the years. It is likely, however, that ammonia concentrations will increase due to climate change, e.g., dryer and warmer springs/summers.

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