



Modelling the future distribution of ammonium nitrate concentrations in The Netherlands for 2020: The sensitivity to meteorological parameters



J.E. Williams^{a, b, *}, E. van der Swaluw^b, W.J. de Vries^b, F.J. Sauter^b, W.A.J. van Pul^b, R. Hoogerbrugge^b

^a Royal Netherlands Meteorological Institute, Utrechtseweg 297, 3731 GA, De Bilt, The Netherlands

^b National Institute for Public Health and The Environment, Centre for Environmental Monitoring, P.O. Box 1, 3720 BA Bilthoven, The Netherlands

HIGHLIGHTS

- We develop an approach for simulating $[\text{NH}_4^+]$ in source-receptor models.
- We apply in the OPS model for the Netherlands.
- Validation shows an accuracy in the annual mean $[\text{NH}_4^+]$ of ~85%.
- Future simulations (2020) show $[\text{NH}_4^+]$ decreases of ~5–25%.
- Uncertainty of ~100% introduced by choice of meteorological fields.

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ABSTRACT

We present a parameterization developed to simulate Ammonium particle (NH_4^+) concentrations in the Operational Priority Substances (OPS) source-receptor model, without the necessity of using a detailed chemical scheme. By using the ratios of the main pre-cursor gases SO_2 , NO_2 and NH_3 , and utilising calculations performed using a chemical box-model, we show that the parameterization can simulate annual mean NH_4^+ concentration fields to within ~15% of measured values at locations throughout the Netherlands. Performing simulations for different decades, we find a strong correlation of simulated NH_4^+ distributions for both past (1993–1995) and present (2009–2012) time periods. Although the total concentration of NH_4^+ has decreased over the period, we find that the fraction of NH_4^+ transported into the Netherlands has increased from around 40% in the past to 50% for present-day. This is due to the variable efficiency of mitigation practises across economic sectors. Performing simulations for the year 2020 using associated emission estimates, we show that there are generally decreases of ~8–25% compared to present day concentrations. By altering the meteorological fields applied in the future simulations, we show that a significant uncertainty of between ~50 and 100% exists on this estimated NH_4^+ distribution as a result of variability in the temperature dependent emission terms and relative humidity. Therefore, any projections of future NH_4^+ distributions should be performed using well chosen meteorological fields representing recent meteorological situations.

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1. Introduction

The concentration of Secondary Inorganic Aerosol (SIA) particles containing ammonium (i.e. ammonium bi-sulphate (NH_4HSO_4),

ammonium sulphate ($(\text{NH}_4)_2\text{SO}_4$) and ammonium nitrate (NH_4NO_3) affects air quality, visibility and, ultimately, the acidification and eutrophication of sensitive ecosystems and surface waters via deposition (Bobbink et al., 1998; Stevens et al., 2010). These particles are formed from the chemical oxidation and subsequent aggregation of pre-cursor gases such as ammonia (NH_3), nitrogen dioxide (NO_2) and sulphur dioxide (SO_2) as a function of both temperature (T) and relative humidity (RH). In Europe, there

* Corresponding author. Royal Netherlands Meteorological Institute, Utrechtseweg 297, 3731 GA, De Bilt, The Netherlands.

E-mail address: williams@knmi.nl (J.E. Williams).

has been a steady decline in resident SO₂ concentrations, and to a lesser extent NO₂ concentrations, over the last few decades (Tørseth et al., 2012). This has resulted in the speciation of particulate ammonium (NH₄⁺) being shifted away from the more stable forms of NH₄HSO₄ and (NH₄)₂SO₄, towards the less stable form of NH₄NO₃, which may undergo evaporation back towards the precursor gases of nitric acid (HNO₃) and NH₃, depending on the meteorological conditions (T, RH). The contribution of NH₄⁺ towards the total mass of SIA in Europe is of the order of 10–20% (Schaap et al., 2004).

In the Netherlands, the sectors responsible for the highest emissions of the pre-cursor gases varies with respect to the chemical species, with nitrogen oxides (NO_x) predominantly originating from transport, NH₃ from agricultural practices and SO₂ from shipping and energy production (for examples see www.emissieregistratie.nl). Therefore, the introduction of different mitigation practices across sectors and changes in the growth of each respective sector over time has resulted in a change in the emission ratios between the respective pre-cursor gases, and thus the efficiency of SIA formation. Although NH₃ emissions in Northern Europe are thought to have been reduced over the last decade (Skjøth and Geels, 2013), the threshold of the critical loads defined for acidification and eutrophication are still frequently exceeded (Beijk et al., 2009). This is in spite of a significant decrease in the wet deposition of NH₄⁺, nitrate (NO₃⁻) and sulphate (SO₄²⁻) over the past few decades (van der Swaluw et al., 2011). Simulations have indicated that ~65% of NH₄⁺ particles are transported into the Netherlands from neighbouring countries (Hendriks et al., 2013), meaning that to achieve National Air Quality targets reductions must be made at European scale. Future emission estimates suggest that the further reduction in NO_x emissions will most likely continue throughout the 21st century. For NH₃, emissions are strongly linked to regional temperatures and the intensification and methodology of agricultural practices (Skjøth and Geels, 2013), with a potential increase by ~50% at global scale by 2100 (Sutton et al., 2013). However, non-linearities inherent in the formation of SIA can lead to rather muted reductions in SIA concentrations, in spite of significant reduction associated with the pre-cursor emissions (Harrison et al., 2013), as has been found at both regional (Fagerli and Aas, 2008) and global scales (Hauglustaine et al., 2014). Therefore, assessing the changes in the concentration of NH₄⁺ ([NH₄⁺]) in the Netherlands when accounting for future mitigation strategies for the near future is warranted.

In this paper we investigate the potential changes in [NH₄⁺] for the Netherlands for time-slices across past and future decades as simulated using the Operational Priority Substances model (OPS). In Sect. 2, we describe the methodology used for deriving a parameterization for calculating NH₄⁺ particle formation in OPS using output taken from simulations performed with a chemical-box model including SO₄²⁻ and NO₃⁻ particle formation, as well as introducing the various sensitivity simulations that are used for this study. In Sect. 3 we show the performance of the new NH₄⁺ parameterization when compared to both historical (1990s) and present-day (2009–2012) measurements taken throughout the Netherlands. In Sect. 4 we show simulations performed for 2020 using state-of-the-art emission estimates and examine the expected changes in the temporal distribution of NH₄⁺ for the coming decade. Finally, in Sect. 5, we present our conclusions.

2. Methodology

2.1. The OPS model

Here we use the OPS source-receptor model, which accounts for the long-range transport, chemical formation and deposition of SIA.

The OPS model represents a combination of a Gaussian plume model, for local-scale application, and a trajectory model for long-range transport (van Jaarsveld and de Leeuw, 1993; van Jaarsveld, 2004). OPS has been applied for a number of previous studies concerned with SIA (e.g. van Pul et al., 2004; Velders and Diederer, 2009; van der Swaluw et al., 2011). The model is driven with yearly-specific meteorological fields provided by the Royal Netherlands Meteorological Institute (KNMI), where values are binned into distinct meteorological stability classes within OPS (van Jaarsveld et al., 2004). Due to the large number of Lagrangian trajectories employed in OPS, it is unrealistic to be able to model a full description of tropospheric chemical processes online and preserve the rather efficient runtime. Therefore, no explicit chemical scheme is included and, thus, parameterizations are employed within OPS for simulating the formation of SIA particles.

For NH₄⁺ we develop a new parameterization using a chemical box-model to describe the formation of NH₄HSO₄, (NH₄)₂SO₄ and NH₄NO₃, as a function of the ratio of the three most important precursor gases (NH₃, SO₂ and NO₂), hereafter referred to as NH₃-CONV. The rate of particle formation is subsequently converted into a pseudo first-order formation rate expressed in terms of percent conversion of NH₃ per hour (%/hr). The OPS model accounts for the effect of temperature on the emission of NH₃ from agricultural sources, where fluxes approximately double for every 5 °C temperature increase as shown in Sutton et al. (2013). For NO_x and SO₂ there is no corresponding temperature feedback on the emission fluxes as these species are predominantly released from anthropogenic sources such as e.g. road transportation and energy production, with biogenic NO_x considered to be a relatively unimportant NO_x source for Europe (Simpson, 1995).

The OPS model also uses annual mean background concentration maps for defining the temporal distribution of the precursor species (and thus ratios) throughout the Netherlands at a 1 by 1 km resolution for specific years (e.g. 1995, 2005) based on interpolated measurements. These background concentrations are needed to constrain the chemical calculations performed in OPS. Interpolation for interstitial years means that the values used are yearly-specific. Fig. 1 shows the temporal distribution of both [NO₂]/[NH₃] and [SO₂]/[NH₃] (hereafter referred to as the C₁ and C₂ ratios) throughout the Netherlands for the years 1994, 2005 and 2020, respectively. The variability for each respective ratio for each individual year is provided in the inset, along with the number of incidences that occur in each bin. These ratios are calculated using the temporal distribution of each pre-cursor species defined in background maps based on an iterative set of simulations using OPS, and subsequently bias corrected using measurements taken throughout the Netherlands. The 1 by 1 km grid is then filled by interpolating values of such bias corrections to fill the whole domain.

The distribution in the range of each of the ratios generally follows the variability in the regional population density and industrial/agricultural activity, with higher NO_x emissions towards the west and higher NH₃ emissions towards the east. This results in a west-east gradient in values, where higher C₁ and C₂ ratios (ranging between 10 and 30) occur around industrial and coastal regions due to relatively low [NH₃]. It should be noted that these background maps were derived using simulations that did not include oceanic emissions of NH₃, although their influence on resident [NH₃] is implicitly included during the bias correction. The decrease in the maximum values of C₁ and C₂ across the timeline provides some indication as to the extent of mitigation of both NO_x and SO₂ emissions in the Netherlands over the decade. These ratios are then subsequently used in determining the rate of NH₄⁺ particle formation in NH₃-CONV. For a more comprehensive description of the OPS model, including details of the meteorology, emissions and

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