Validation of satellite-constrained ammonia using a CTM and ground measurements

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INTRODUCTION

Ammonia (NH3) has received a lot of attention nowadays due to its impact on the regulation and the emissions from Earth’s radiative balance. Sources of ammonia include wild animals, wildfires, sewage, systems, humans, biomass burning, volcanic eruptions and agriculture (Eriksen et al., 2007) (Figure 1). The latter is responsible for the largest atmospheric emissions of ammonia. Emissions have increased considerably since pre-industrial times and are unlikely to decrease due to the growing demand for food and feed (Aneja et al., 2008).

Sources of ammonia

(Dentener & Crutzen, 1994)


Figure 1. Primary global sources of ammonia.

Despite its importance, ammonia is one of the most poorly quantified trace gases, with errors over 50% on the global emission budget and even higher on regional and local scales (Dentener and Crutzen, 1994). In the present study we use 2008-2017 of satellite measurements of ammonia retrieved from MASI to calculate surface emissions. The calculated emissions are then compared to the Chemistry Transport Model (CTM) and simulated for the same 10-year period. To verify the improvement of the calculated emissions of ammonia, we evaluate the modelled surface concentrations against ground-based measurements from different monitoring stations. The same comparison is performed for the most recent state-of-the-art emission datasets.

FLUX CALCULATIONS OF AMMONIA

Emission fluxes of ammonia were calculated using a 1-dimensional box model that assumes first-order loss terms for ammonia (Van Damme et al., 2018). It takes into account the grid size of the boxes and the temporal resolution of ammonia in a hypothetical atmospheric box following the next equation:

\[ \frac{dN}{dt} = -k * N \]

where \( N \) is the mass of ammonia in each grid cell (in molecules cm\(^{-2}\)) and \( k \) is the lifetime of the ammonia in the box (given in seconds). The MASI measurements were set into a 2-dimensional grid of 0.7°×0.5° using an Inverse Distance Weighting (IDW) method (Ronkai, 2015) preferred for its ease of use and high quality of interpolation.

RESULTS & DISCUSSIONS

Life-time of ammonia is in the order of a few hours to a few days (Behera et al., 2013), thus ammonia can be only transported over short distances as (a) the majority of its emissions occur at the surface, (b) it is highly dependent on deposition velocities over surfaces (Hov et al., 1994). Lifetime depends on numerous factors such as the presence of ammonia’s reactants, atmospheric water vapor and temperature, which vary during daytime and nighttime emissions. At ammonia-poor conditions, ammonia is rapidly removed by neutralizing sulfatic acid. If ammonia increases further above saturation, it may react with nitric acid to form ammonium nitric acid if sulfate concentrations decrease, then free ammonia is produced, which gradually reacts with nitric acid and producing aerosol phase ammonia. In most cases, ammonia is known to have a short lifetime due to its volatility and small size, with a half-life of 20 minutes. However, in some regions, it can last longer, with a half-life of 2 hours.

Figure 2. Lifetime of NH3 (hours).

The lifetime of ammonia is highly variable, due to the chemical loss and deposition processes that affect it. To tackle this problem, a technique called FLUX is used. FLUX is an in-situ measurement of a constant value, so we can calculate emissions from a CTM. This gives robustness in the calculated fluxes; e.g., at regions where sulfatic and nitric acids are abundant, chemical loss will be more efficient and, thus, lifetime much shorter affecting the fluxes. In a CTM, lifetime of ammonia can be easily calculated by the species mass balance equation (O’Neill et al., 2014):

\[ \frac{dC(t)}{dt} = r(t) - \alpha C(t) \]

where \( C(t) \) is the atmospheric burden of ammonia at time \( t \) and \( r(t) \) is the time-dependent source emissions and \( \alpha \) is the removal timescale. Assuming steady-state conditions and continuous fluxes, there is a quasi-equilibrium between sources and removals of ammonia and the modelled aerosol lifetime \( \tau_{mod} \) can be written as:

\[ \tau_{mod} = \frac{C_{NH3}}{r_{trans.chem.dep}} \]

where \( C_{NH3} \) is the atmospheric burden of ammonia and \( r_{trans.chem.dep} \) is the total loss due to any process affecting ammonia in the model (transport, chemical loss, dry deposition).

REFERENCES


Figure 2. FLUX calculations of ammonia.