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Implementation of different big-leaf canopy reduction functions in the Biogenic Emission Inventory System (BEIS) and their impact on concentrations of oxidized nitrogen species in northern Europe

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ABSTRACT

Canopy reduction describes NO_2 flux reduction at leaf stomata. We implemented the big-leaf reduction approaches of Wang et al. (1998) and Yienger and Levy (1995) in the Biogenic Emission Inventory System (BEIS) and compared them with the BEIS standard approach. The different reduction functions lead to a reduction of 17 Gg N or 27 Gg N respectively of nitrogen emission in comparison to the standard approach which reduces the nitrogen flux by about 1 Gg N in the three summer months of 2012. These are significant differences to the standard approach. The concentration reduction of oxidized reactive nitrogen in the model area shows also a significant reduction. While concentration reduction in central europe is low, in more rural regions of Europe, concentration changes are considerably higher. The calculated concentrations of NO_2 show a significant improvement of the model performance when compared to EMEP observations in central Europe. This study favors the implementation and use of canopy reduction factors, especially the parameterization of Wang et al. (1998), for regional and global emission models for reasons of model physical correctness and improved model results.

1. Introduction

Natural emissions of oxidized nitrogen are of lower magnitude than anthropogenic emissions of oxidized nitrogen (Vinken et al., 2014). However, anthropogenic emissions have decreased and are expected to decrease in Europe in the coming decades so that natural and semi-natural emissions will become more and more important for the nitrogen budget. Therefore the contribution of oxidized nitrogen with a natural origin to the total budget and the spatial distribution of natural emissions and contribution to the local budget is a scientifically rewardable question to be addressed.

The formation of different nitrogen species and therefore the concentration and deposition patterns of these species is dominated by nitrogen monoxide and nitrogen dioxide (Derwent et al., 1998). Oxidized nitrogen is a major part of reactive nitrogen. Reactive nitrogen is of fundamental importance for all life processes. If the intake of reactive nitrogen exceeds the eco-system dependent needed nutrient load, ecosystem change or damage may happen.

Nitrogen oxides play a major role in tropospheric chemistry (Crutzen, 1979). They are driving the ozone cycle and are involved in the acidification of rainwater due to the formation of nitric acid in cloud droplets. Nitrogen monoxide and nitrogen dioxide are mainly emitted by anthropogenic combustion processes. Other sources are lightning and to a smaller degree agricultural activities. The biggest natural source of atmospheric nitrogen oxides is the biogenic emission of nitrogen monoxide from soil. Nitrification and denitrification in microorganisms in soil account for 15% of the total global nitrogen oxide emissions (Vinken et al., 2014; Davidson and Kingerlee, 1997; Yienger and Levy, 1995).

The aim of this study is to asses the importance of vegetation as a sink for natural nitrogen oxide emissions. Immediately after the emission and release from soil, a part of the nitrogen monoxide is oxidized to nitrogen dioxide (S. Bakwin et al., 1990; Jacob and Bakwin, 1991; Yienger and Levy, 1995). On its way into the free atmosphere, the flux of nitrogen dioxide is reduced by the absorption of nitrogen dioxide at the stomatal openings of leafs ((Rogers et al., 1979; Jacob and Bakwin, 1991)). This effect is called canopy reduction and is a natural sink for primary biogenic emission of nitrogen.

At Helmholtz-Zentrum Geesthacht, Institute for Coastal Research the Chemistry Transport Model CMAQ with the Sparse Matrix Operator Kernel Emission model SMOKE for Europe (SMOKE-EU) and the Biogenic Emission Inventory System (BEIS) is used for computing biogenic nitrogen oxide soil emissions and its transport and chemical transformation in the atmosphere. For canopy reduction, BEIS uses a

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simple time-dependent assumption. In this study two alternative parameterizations were tested and the impact of the three implementations on the emission and concentration patterns in northern Europe for the Summer of 2012 were investigated.

2. Biogenic emissions and canopy reduction

2.1. Biogenic NO emission from soil in BEIS 3.14

Microorganisms in soil utilize nitrogen as a main nutrient for their metabolic processes. The processes of denitrification (equation (1)) and nitrification (equation (2)) occured in this metabolic processes and release gaseous nitrogen monoxide. This nitrogen monoxide exhausts from the soil surface to the atmosphere.

$$NH_3 \rightarrow NH_2OH \rightarrow NO_2^- \rightarrow NO_3^- \quad (1)$$

$$NO_3^- \rightarrow NO_2^- \rightarrow \mathbf{NO} \rightarrow N_2O \rightarrow N_2 \quad (2)$$

Microbial activity is highly related to the soil temperature and soil moisture. Yienger and Levy (1995) describes, based on Williams and Fehsenfeld (1991), a temperature-, soil moisture-, time- and landuse-dependend soil NO flux parameterization. This parameterization is used in the BEIS model. In the model it is splitted in an landuse-dependend emission factor and a correction factor. The correction factor is constructed following equation (1), 7b and 9a in Yienger and Levy (1995).

After rain events, soil moisture is increased and microbial activity is gained. This is called pulsing. In BEIS the pulsing of NO emissions after rain events is parameterized following equations (4)–(6) of Yienger and Levy (1995). Fertilization effect is simply parameterized by a declining function, where the fertilization effect leads to no emission reduction in the first 30 days of the growing season and after day 30 for the remaining 184 a decline from 0% reduction to 100% reduction of the NO emission of agricultural used soils.

Nitrogen Oxides are absorbed and reemitted by the stomatal openings of leafs, depending of the ambient air concentration. This is called bi-directional flux. BEIS does not include a bi-directional flux scheme, only absorbtion is parameterized (see 3.1).

2.2. General canopy reduction process

Canopy Reduction describes the uptake of nitrogen dioxide gas at the stomatal opening of leafs and the resulting reduction of nitrogen dioxide flux through the canopy (Yienger and Levy, 1995; Wang et al., 1998; Ganzeveld et al., 2002; Rasool et al., 2016). The gaseous nitrogen dioxide dissolves in the water belonging to the stomata and gets incorporated by the plant (Rogers et al., 1979). This reduces the flux of nitrogen oxides from the soil to the atmosphere. The worldwide reduction of nitrogen dioxide emissions resulting from canopy reduction is in the order of 20%. Local amounts of reduction can be as large as 70% (e.g. in the tropic rain-forest region) (Wang et al., 1998).

It is important to keep this in mind that despite the fact that biogenic emissions are mainly nitrogen monoxide emissions, it is the flux of nitrogen dioxide to the atmosphere which is reduced. Nitrogen dioxide is about ten times more water soluble than nitrogen monoxide. This makes nitrogen dioxide flux reduction on leafs considerably more efficient. Because nitrogen monoxide is highly reactive, it is assumed that a certain amount of nitrogen monoxide is already oxidized to nitrogen dioxide when it reaches the canopy (S. Bakwin et al., 1990; Jacob and Bakwin, 1991; Yienger and Levy, 1995). Canopy reduction models use typically a fixed fraction to calculate the reduction amount in terms of primary emitted nitrogen monoxide. A typical NO_2 to NO_x split in the canopy is 0.5–0.9 (Jacob and Bakwin, 1991), with 0.7 as used in Wang et al. (1998).

All three tested parameterizations are big-leaf approaches. This means that only one big leaf is assumed as canopy. In contrast, multi-layer approaches consider multiple leaf layers in more complex vegetation-atmosphere exchange models. For the polluted region of Europe big-leaf approaches are suitable (see Ganzeveld et al. (2002)) and technically most efficient because of their simplicity in implementation. In more complex soil models, big-leaf approaches are also used. Implemented, but not fully tested and evaluated was the Wang et al. (1998) parameterization in Rasool et al. (2016) as a part of an updated soil NO emission model BDSNP for the CMAQ model system.

It should be noted that canopy reduction does not only affect natural emissions from soil. Anthropogenic emissions in rural areas might flow through the canopy first by horizontal advection and afterwards by mixing with higher atmospheric layers. Thus, anthropogenic emissions may also be affected, but there is no previous research and literature about this effect.

3. Different canopy reduction schemes used in this study

3.1. Current implementation in emission model BEIS 3.14: Vukovich and Pierce (2002)

The Biogenic Emission Inventory System (BEIS) (Vukovich and Pierce, 2002) models biogenic emissions of nitrogen oxide with a soil temperature dependent function and a classification of agricultural, non-agricultural and growing-season or non-growing-season of the underlying land use class. For the "agricultural growing season" case, there is a vegetation adjustment factor (a model-internal synonymous for the (1-CRF) canopy reduction factor formulation) that scales between 1 and 0.5, depending on the day of the growing season. Furthermore, the vegetation adjustment factor is only applied to one of three possible cases. The first possible case is frozen soil, where no NO is emitted. The second is the calculation of grassland emissions with no adjustments for vegetation, rain or fertilization. The third one is the plant specific emission with adjustment for vegetation, rain and fertilizer. The vegetation adjustment is only used, when the soil is not frozen and the pure grassland emission is lower than the plant specific emission. This is an incomplete approximation. In particular, it neglects the reduction of non-agricultural emissions in more natural landscapes.

The original description of the BEIS model in the SMOKE framework does not include the vegetation adjustment, the parameterization with the vegetation factor is mentioned in Pouliot and Pierce (2009) and according to the CMAS model history implemented directly in the model version described in Vukovich and Pierce (2002). Therefore, the original implementation of the parameterization is denoted here as **VP02**.

3.2. Parameterization of Yienger and Levy (1995)

Yienger and Levy (1995) describe different influence factors for nitrogen oxide emissions from soil. Their aim is an emission estimate for global annual nitrogen oxide from soil. They use gridded geographic and geoscientific information like the land use class and temperature as input. One of the influence factors for the emission estimate is the canopy reduction for which they developed a parameterization. It is based on the leaf area index (LAI) and the stomatal area index (SAI). Both are parameterized through the gridded land use class.

$$CRF = \frac{exp(-k_s \times SAI) + exp(-k_c \times LAI)}{2} \quad (3)$$

with *CRF* as Canopy Reduction Factor, $k_s = 8.75\ m^2\ m^{-2}$ and $k_c = 0.24\ m^2\ m^{-2}$ as absorbtion coefficients.

3.2.1. Model limitation and adaption measures

The model is a gray absorber model and has some limitations. The vertical transfer and therefore the canopy residence time is not taken into account. The model does not contain a stomata resistance model either. As described above, the emissions and influence factor