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# CRITICAL LOADS OF OXIDIZED SULPHUR, OXIDIZED AND NUTRIENT NITROGEN

## 2015

*This report presents recent results of the calculations of critical loads and exceedances of nitrogen and sulphur compounds in Lithuania*

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# CRITICAL LOADS OF OXIDIZED SULPHUR, OXIDIZED AND NUTRIENT NITROGEN

## INTRODUCTION

This report presents recent results of the calculations of critical loads and exceedances of nitrogen and sulphur compounds in Lithuania.

Changes in air concentrations and depositions from year to year are driven by changes in both emissions and meteorology. Between 2013 and 2014, emissions of  $\text{SO}_x$ ,  $\text{NO}_x$  and  $\text{NH}_3$  in the extended EMEP area decreased by 0.1 %, 2.0 % and 0.2 %, respectively. Individual countries have reported much larger changes. 2014 was a relatively dry year in large parts of central, south-western and south-eastern Europe, especially in warm season. In these areas, depositions slightly decreased. In Lithuania 2014 was less emissions, in this reason critical loads was slightly less than in 2013. The calculated exceedances of critical loads and the ecosystem areas at risk in 2014 are presented in Figure 3 in the EMEP domain. In total, depositions of sulphur has decreased by ~0.4 %, whilst deposition of oxidized nitrogen and reduced nitrogen have decreased by 2 % and 0.4 %, respectively, which is very similar to the emission changes.

Between 2013 and 2014, emissions of  $\text{SO}_x$ ,  $\text{NO}_x$  and  $\text{NH}_3$  in the extended EMEP area decreased with 0.1 %, 2.0 % and 0.2 %, respectively. Due to decrease of emissions alone (excluding the meteorological variability), there were little change in sulphur deposition. Meteorological conditions have a significant effect on air concentrations and depositions of pollutants, controlling their transport, diffusion and dry and wet removal.

## METHODS AND DATA SOURCES

The starting point for calculating critical loads of nutrient N by the SMB (Simple Mass Balance) model is the mass balance of total nitrogen for the soil compartment under consideration (inputs=sinks+outputs):

**Equation 1**

$$N_{dep} + N_{fix} = N_{AD} + N_i + N_u + N_{de} + N_{eros} + N_{fire} + N_{vol} + N_{le};$$

where  $N_{dep}$  is the total N deposition,  $N_{fix}$  is the N “input” by biological fixation,  $N_{ad}$  is N adsorption,  $N_i$  is the long-term net immobilization of N in soil organic matter,  $N_u$  is the net removal of N harvested vegetation and animals,  $N_{de}$  is flux of N to the atmosphere due to denitrification,  $N_{eros}$  are N losses through erosion,  $N_{fire}$  are N losses in smoke due to (wild or controlled) fires to the atmosphere,  $N_{vol}$  are N losses to the atmosphere via  $NH_3$  volatilization, and  $N_{le}$  is leaching of N below the root zone.

The following assumptions lead to a simplification of Eq. (1): nitrogen adsorption, e. g., the adsorption of  $NH_4$  by clay minerals, can temporarily lead to an accumulation of N in the soil, however it is stored/released only when the deposition changes, and can thus be neglected in steady-state considerations; nitrogen fixation is negligible in most (forest) ecosystems, except for N-fixing species; the loss of N due to fires, erosion, and volatilization is small for most ecosystems in Europe, and therefore neglected. Alternatively, one could replace  $N_i$  by  $N_i + N_{eros} + N_{fire} + N_{vol} - N_{fix}$  in the subsequent equations. The leaching of ammonium ( $NH_4$ ) can be neglected in all forest ecosystems due to (preferential) uptake and complete nitrification within the root zone (i.e.,  $NH_{4,le} = 0$ ,  $N_{le} = NO_{3,le}$ ). Under these simplifying assumptions Eq. (1) becomes:

**Equation 2**

$$N_{dep} = N_i + N_u + N_{de} + N_{le}.$$

From this equation a critical load is obtained by defining an acceptable limit to the leaching of N,  $N_{le(acc)}$ , the choice of this limit depending on the ‘sensitive element of the environment’ to be protected. If an acceptable leaching is inserted into Eq. (2), the deposition of N becomes the critical load of nutrient nitrogen,  $CL_{nut}(N)$ :

**Equation 3**

$$CL_{nut}(N) = N_i + N_u + N_{de} + N_{le(acc)}.$$

In deriving the critical load of nutrient N as Eq. (3), it is assumed that the sources and sinks do not depend on the deposition of N. This is unlikely to be the case and thus all quantities should be taken 'at critical load'. However, to compute, e.g., 'denitrification at critical load' one needs to know the CL, the very quantity one wants to compute. The only clean way to avoid this circular reasoning is to establish a functional relationship between deposition and the sink of N, insert this function into Eq. (2) and solve for the deposition (to obtain the critical load). This has been done for denitrification: in the simplest case denitrification is linearly related to the net input of N (De Vries et al., 1993, 1994):

**Equation 4**

$$N_{de} = \begin{cases} f_{de} \cdot (N_{dep} - N_i - N_u) & \text{if } N_{dep} > N_i + N_u \\ 0 & \text{else} \end{cases},$$

where  $f_{de}$  ( $0 < f_{de} < 1$ ) is the so-called denitrification fraction, a site-specific quantity. This formulation implicitly assumes that immobilization and uptake are faster processes than denitrification. Inserting this expression for  $N_{de}$  into Eq. (2) and solving for the deposition leads to the following expression for the critical load of nutrient N:

**Equation 5**

$$CL_{nut}(N) = N_i + N_u + \frac{N_{le(acc)}}{1 - f_{de}}.$$

The acceptable N leaching (in eq/ha/yr) is calculated as:

**Equation 6**

$$N_{le(acc)} = Q \cdot [N]_{acc}$$

where  $[N]_{acc}$  is the acceptable N concentration (eq/m<sup>3</sup>) and Q is the precipitation surplus (in m<sup>3</sup>/ha/yr). Values for acceptable N concentration are given in Table 1 (De Vries et al., 2007).

**Table 0-1 Critical (acceptable) N concentrations in soil solution for calculating  $Cl_{nut}(N)$  (De Vries et al., 2007).**

Impact	$[N]_{acc}$ , mgN/L
Vegetation changes (data established in the Netherlands) <sup>1</sup> :	
Coniferous forest	2.5–4.0
Deciduous forest	3.5–6.5
Grass lands	3.0
Heath lands	3.0–6.0
Other impacts on forests:	
Nutrient imbalances	0.2–0.4
Elevated nitrogen leaching/N saturation	1.0
Fine root biomass/root length	1.0–3.0
Sensitivity to frost and fungal diseases	3.0–5.0

<sup>1</sup>Note that these values should be used with caution, e.g., in areas with high precipitation.

Dutch and Ineson (1990) reviewed data on rates of denitrification. Typical values of  $N_{de}$  for boreal and temperate ecosystems are in the range of 0.1–3.0 kgN/ha/yr (=7.1–214.3 eq/ha/yr), where the higher values apply to wet(ter) soils; rates for well drained soils are generally below 0.5 kgN/ha/yr.

The long-term annual N immobilization of nitrogen was set to 0.2–0.5 kgN/ha/yr (14.3–35710 eq/ha/yr). Considering that the immobilization of N is probably higher in warmer climates, values of up to 1 kgN/ha/yr (71.4 eq/ha/yr) could be used for  $N_i$ , without causing unsustainable accumulation of N in the soil.

Critical loads of S,  $CL(S)$ , and N,  $CL(N)$ , can be computed by defining a critical ANC leaching,  $ANC_{le}$ :

**Equation 7**

$$CL(S) + CL(N) = BC_{dep}^* - Cl_{dep}^* + BC_w - Bc_u + N_i + N_u + N_{de} - ANC_{le,crit}$$

where  $BC$  is the sum of base cations, where the subscripts  $w$  and  $u$  stand for weathering and net growth uptake,  $ANC_{le,crit}$  is Acid Neutralizing Capacity.

Critical loads of sulphur and nitrogen, both contributing to acidification of ecosystems, and their exceedances were derived and mapped in a large scale exercise for forest soils (deciduous, coniferous and mixed forest), natural grassland, acidic fens, heathland and mesotrophic peat bogs in Lithuania. Each ecosystem has its specific sensitivity against the air pollutants, which is expressed by the critical load value. To identify this, the geographical information from CORINE land cover database has to be overlapped with spatial information on soil and climate. In combination with the General Soil Map of Lithuania and climate data conclusions on the vegetation structure of the land cover types can be drawn and the net biomass production can be derived.

The EMEP Eulerian acid deposition model output has been used as deposition of nitrogen and sulphur compounds in Lithuania.

Every year, emission data per sector from Lithuania to the LRTAP Convention were compiled at a national level and were reported through the EMEP program. The emission data were reported in the Nomenclature For Reporting (NFR) source categories. There are 120 NFR categories in the reporting templates, including both detailed categories to facilitate reporting under the Convention. The national inventory is based on national statistics and country specific, technology dependent emission factors according to the EMEP/CORINAIR Emission Inventory Guidebook. In addition, new routines and standards for validating emission data have recently been adopted (UNECE, 2005). The background data (activity data and emission factors) for estimation of the Lithuanian emission inventories are collected and stored in databases.

## CRITICAL LOAD AND EXCEEDANCES MAPS

Annual critical loads and total (dry and wet) deposition values of oxidized sulphur, oxidized and nutrient nitrogen were figured on 50×50 km<sup>2</sup> EMEP grid. Critical loads for Lithuania ecosystems were evaluated by using GIS model LandUse. During the evaluation of critical loads the distributions over the territory of Lithuania of coniferous, deciduous and mixed woods, annual average temperature, average annual precipitation and soil map were taken into account.

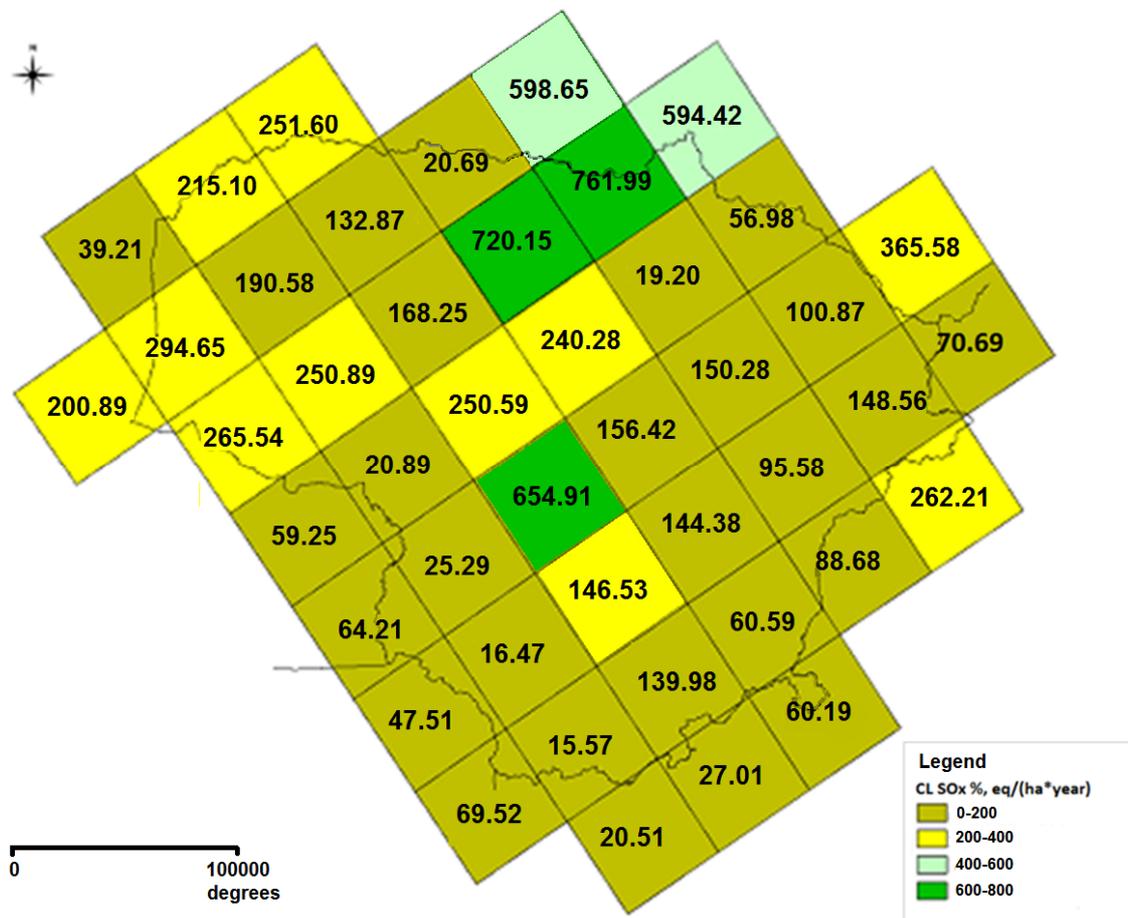


Figure 1 Critical loads of oxidized sulphur (50×50 km<sup>2</sup>), eq•ha<sup>-1</sup>yr<sup>-1</sup>

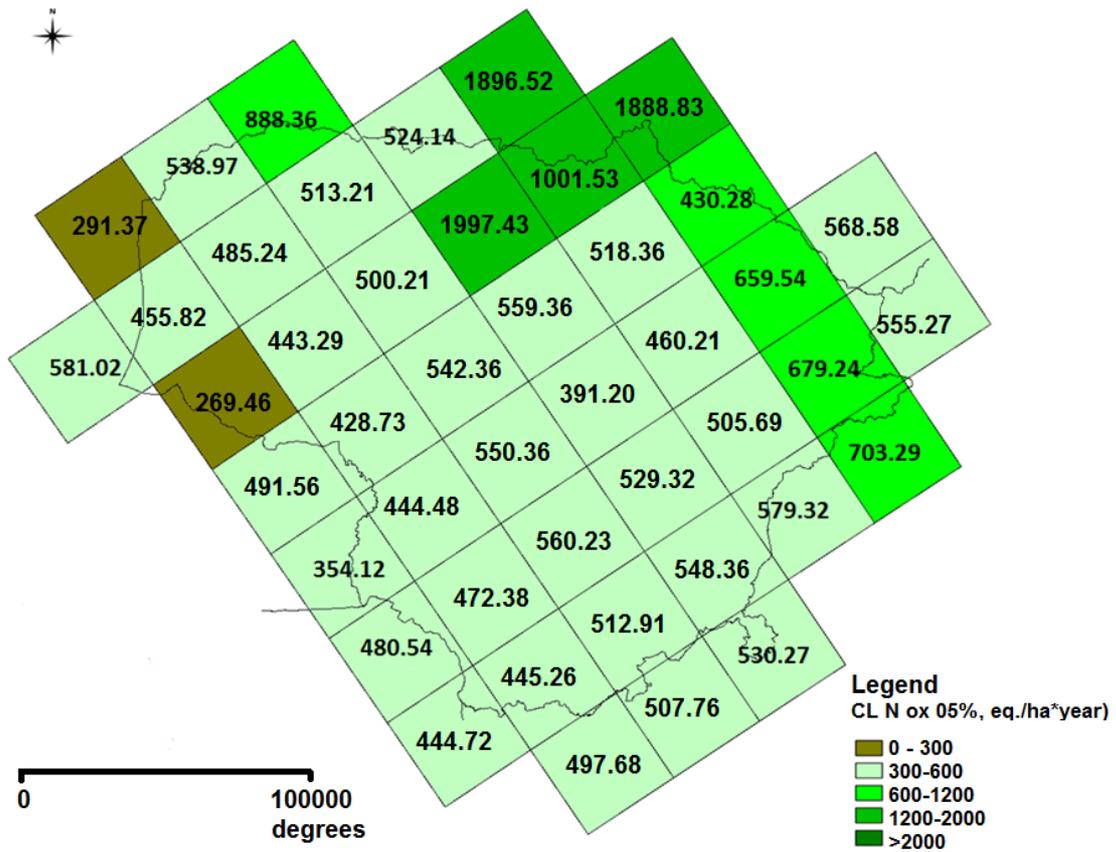


Figure 2 Critical loads of oxidized nitrogen (50×50 km<sup>2</sup>), eq·ha<sup>-1</sup>·yr<sup>-1</sup>.

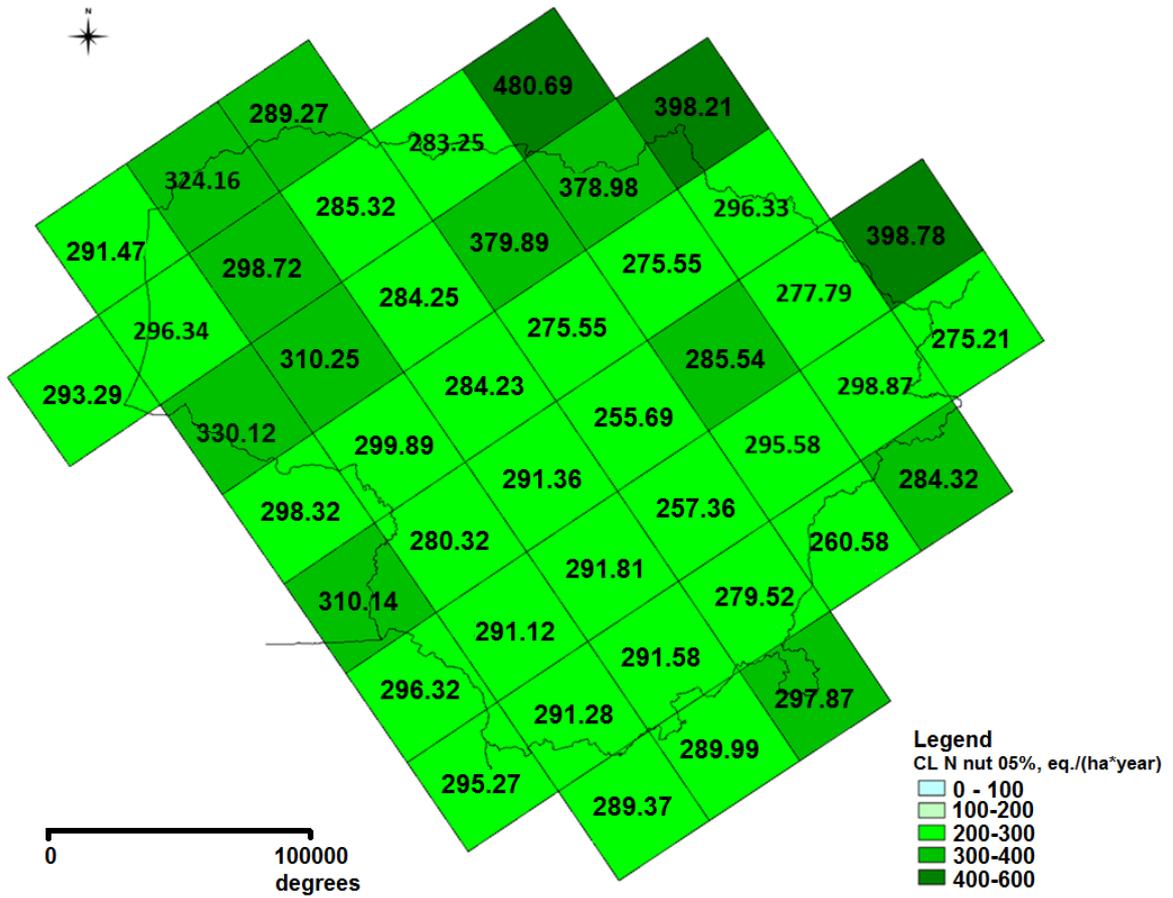
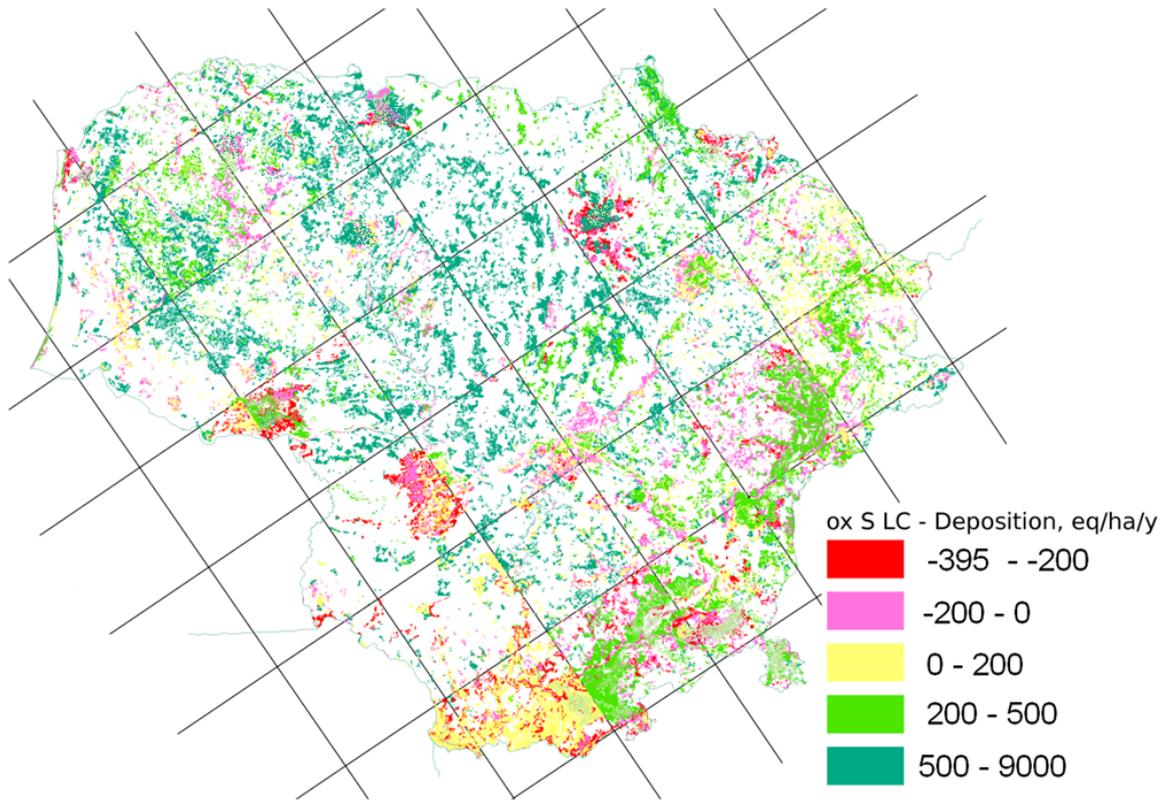
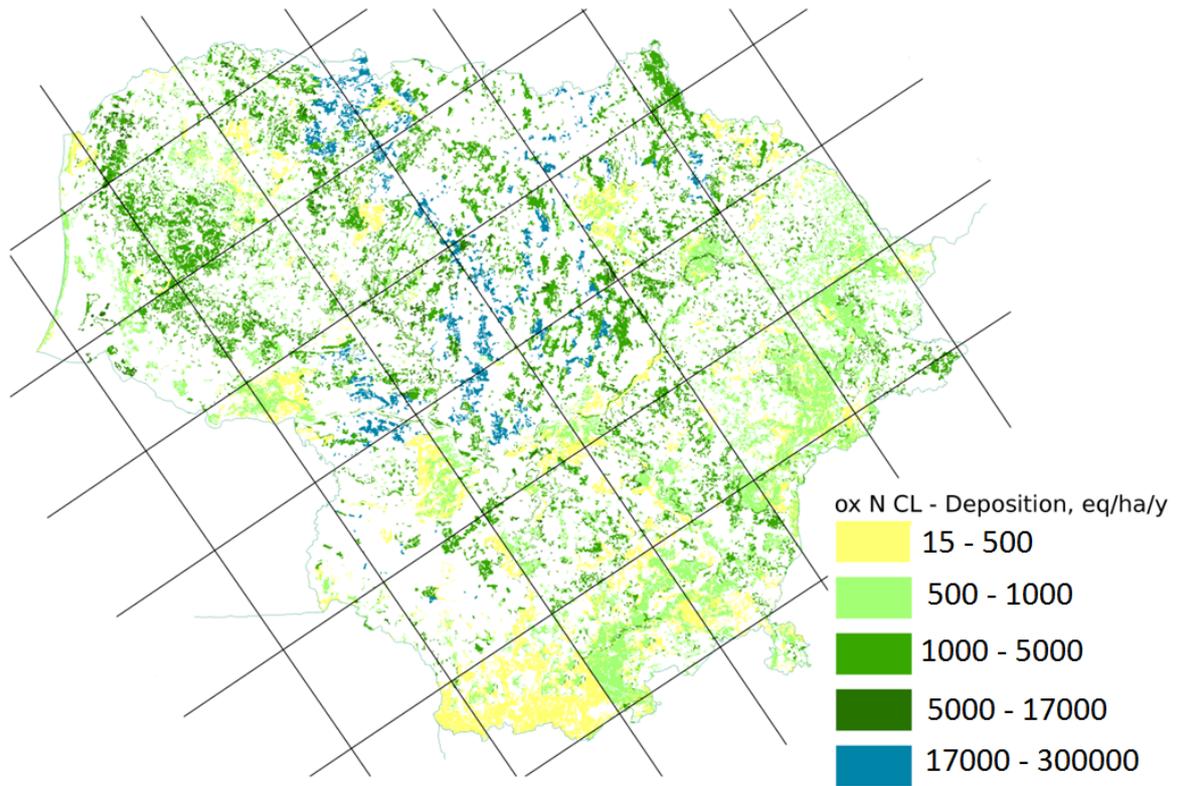


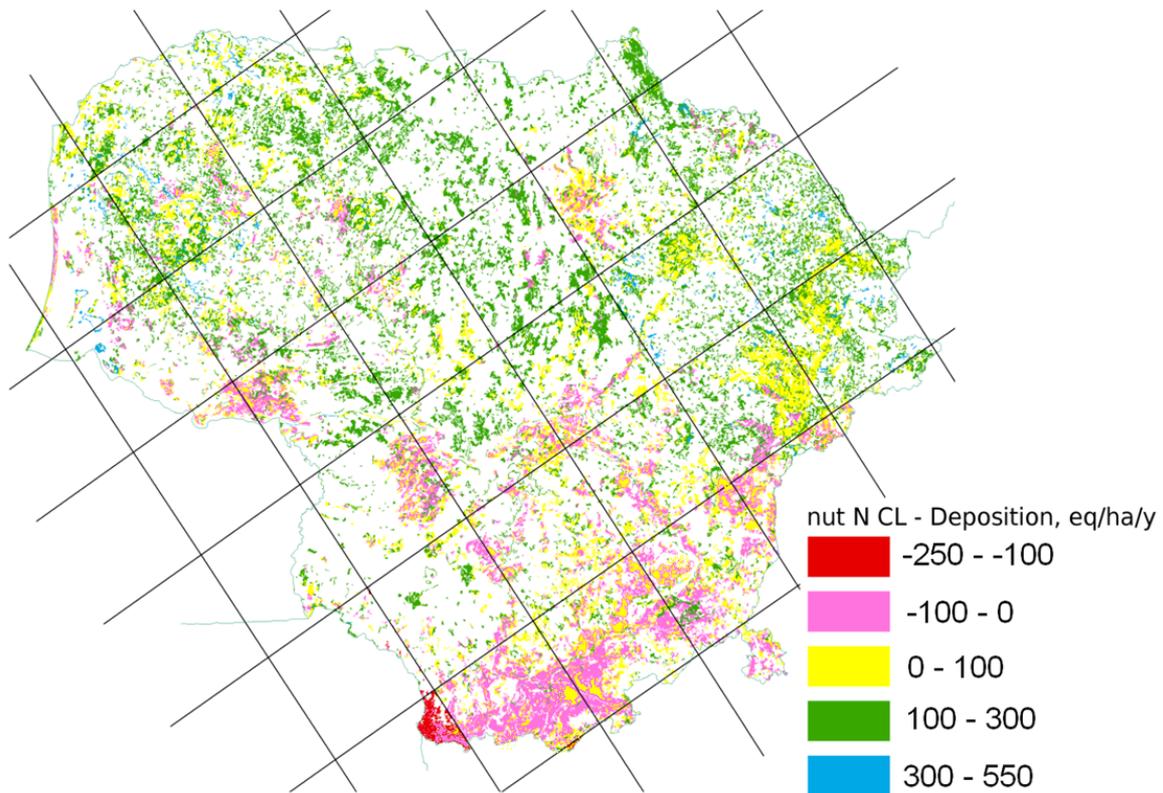
Figure 3 Critical loads of nutrient nitrogen (50x50 km<sup>2</sup>), eq•ha<sup>-1</sup>yr<sup>-1</sup>



**Figure 4** Difference of critical loads and deposition of oxidized sulphur; negative values represent exceedances of critical load (50×50 km<sup>2</sup>), eq·ha<sup>-1</sup>yr<sup>-1</sup>



**Figure 5** Difference of critical loads and deposition of oxidized nitrogen; negative values represent exceedances of critical load (50×50 km<sup>2</sup>), eq·ha<sup>-1</sup>·yr<sup>-1</sup>



**Figure 6 Difference of critical loads and deposition of nutrient nitrogen; negative values represent exceedances of critical load ( $50 \times 50 \text{ km}^2$ ),  $\text{eq} \cdot \text{ha}^{-1} \cdot \text{yr}^{-1}$**

Calculated critical loads values of oxidized sulphur, oxidized and nutrient nitrogen are shown in Figs. 1-3. Calculations of critical loads were made for 5<sup>th</sup> percentile, i.e. 95% of ecosystem can sustain such load. Oxidized sulphur critical load values varied from 15.57 to 761.99  $\text{eq} \cdot \text{ha}^{-1} \cdot \text{yr}^{-1}$  (Fig. 1). The highest critical load values of oxidized sulphur were calculated for the northern and central parts of Lithuania, the lowest – for southern parts.

Critical load values of oxidized nitrogen varied from 269 to 1896  $\text{eq} \cdot \text{ha}^{-1} \cdot \text{yr}^{-1}$  (Fig. 2). The lowest critical load values of oxidized nitrogen were calculated for the southern part of Lithuania.

Critical load values of nutrient nitrogen varied from 261 to 481  $\text{eq} \cdot \text{ha}^{-1} \cdot \text{yr}^{-1}$ . Fig. 3 shows, that the highest critical load values of nutrient nitrogen were calculated for the northern and western parts of Lithuania, and the lowest – for southern parts.

The difference of critical loads and total depositions of oxidized sulphur, oxidized and nutrient nitrogen was calculated, whose negative values represent exceedances of critical load. Due to the time-dependence of atmospheric deposition of pollutants, exceedances are theoretically speaking only valid for a given moment in time (Hettelingh et al., 2009). Consequently the time, for which the exceedances have been calculated, has to be reported. We calculated the exceedances for the deposition data of year 2008, because the newer deposition data were not available.

The calculated differences of critical loads and deposition of oxidized sulphur ( $-395 - 9000 \text{ eq}\cdot\text{ha}^{-1}\text{yr}^{-1}$ ) are shown in the Fig. 4. As can be seen, critical loads of oxidized sulphur were mostly exceeding in the southern, southwestern and small northern parts of Lithuania.

The calculated differences of critical loads and deposition of oxidized nitrogen ( $15 - 27380 \text{ eq}\cdot\text{ha}^{-1}\text{yr}^{-1}$ ) are shown in the Fig. 5. As can be seen, critical loads of oxidized nitrogen were not exceeded over all territory of Lithuania.

The calculated differences of critical loads and deposition of nutrient nitrogen ( $-250 - 550 \text{ eq}\cdot\text{ha}^{-1}\text{yr}^{-1}$ ) are shown in the Fig. 6. As can be seen, the highest exceedances of critical loads of nutrient nitrogen were calculated for the southern part of Lithuania. The lowest exceedances of critical load of nutrient nitrogen were calculated for the northern parts of Lithuania.

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