



**Chemical composition of wet deposits and drainage runoff in agroecosystems:  
the case of Middle Lithuania**

*Angelija Bučienė, Kazimieras Gaigalis*

Bučienė, A., Gaigalis, K., 2012. Chemical composition of wet deposits and drainage runoff in agroecosystems: the case of Middle Lithuania. *Baltica*, 25 (2), 153-162. Vilnius. ISSN 0067-3064.

Revised manuscript submitted 11 September 2012 / Accepted 4 November 2012 / Published online 10 December 2012  
© Baltica 2012

**Abstract** The consistent patterns of chemical composition of wet deposits and drainage runoff concentration have been permanently studied in two stationary sites in the Middle Lithuania for the last decades since late 1980s. The results of simultaneous observations presented in this paper show that the concentration of  $\text{SO}_4^{2-}$ -S,  $\text{Mg}^{+2}$ ,  $\text{PO}_4^{-3}$ -P,  $\text{NO}_3^{-}$ -N,  $\text{NH}_4^{+}$ -N and  $\text{Ca}^{+2}$  in wet deposits positively correlates with the concentration of these ions in drainage runoff water ( $R=0.59, 0.61, 0.51, 0.49, 0.42$  and  $0.35$ , correspondingly,  $R_{05}=0.35$ ). A sharp decrease in  $\text{SO}_4^{2-}$ -S concentration from 5.5 to 2.5  $\text{mg l}^{-1}$  and periodical decrease versus increase in  $\text{NO}_3^{-}$ -N and  $\text{NH}_4^{+}$ -N concentration and load are determined in the agroecosystems studied from 1988–1989 with short intervals to 2007. This outcome has mainly an economic character: impact of changes in animal numbers, increase of transport load, fuel consumption and industrial emissions.

**Keywords** • Wet deposits • Chemical concentration • Drainage runoff • Agroecosystems • Lithuania

✉ Angelija Bučienė [angelija.buciene@ku.lt], Faculty of Social Sciences, Klaipėda University, Minijos 153, LT-93185 Klaipėda; Kazimieras Gaigalis, Water Research Institute, Aleksandras Stulginskis University, Parko 6, Vilainiai, LT-58102 Kedainiai, Lithuania.

## INTRODUCTION

Atmosphere deposits are involved in the water cycle and migration of matter. They can carry and load large inputs of different nutrients which occur on the soil surface or in deeper layers and can leach with drainage runoff to the nearest streams (HELCOM 2002). Rosenberg *et al.* (1990) estimated that about 50% of the nitrogen load on the overall Baltic Sea arises from atmospheric deposition. Lindfors *et al.* (1993) have found during their field studies that dry deposition contributes to between 10 and 30% of the atmospheric nitrogen input to the Baltic Sea (Hertel *et al.* 2003). The history and forecast of the main emissions in Europe has been compiled and calculated by Schöpp *et al.* (2003). The estimations showed that the emissions of acidifying gases in Europe peaked in the 1970s, just before the implementation of the International Protocols. By 2000, sulphur deposition had decreased by >50% and nitrogen by ~20% (Wright *et al.* 2005). Further decreases are predicted for the next 20 years if the Gothenburg Protocol, which sets emission ceilings for 2010 for four pollutants – sulphur,  $\text{NO}_x$ , VOCs

and ammonia – and other national legislations are implemented. Mean nitrogen throughfall deposition ranged from 9.1 to 11.0  $\text{kg ha}^{-1}\text{yr}^{-1}$  measured in 2000–2005 for about 220 plots in Europe. The nitrate deposition ranged from 4.6 to 5.7  $\text{kg ha}^{-1}\text{yr}^{-1}$  (Ulevičius *et al.* 2009).

The periodical analysis of wet precipitation in Lithuania was started since 1980 (Jucevičienė 2003). The impact of precipitation rate on the drainage discharge/runoff and amount of main nutrients leaching is presented and discussed in the scientific papers of many authors (Deelstra *et al.* 2009; Gustafson 1988; Fagerberg *et al.* 1993; Djodjic *et al.* 2004; Tumas 1997 and 1999; Gužys 2002; Gužys, Arlauskienė 2001; Povilaitis 2004 and 2006; Gaigalis *et al.* 2007; Šmitienė *et al.* 2005; Mašauskas *et al.* 2006; Šileika *et al.* 2005; Bučienė *et al.* 2007). However there are too few findings concerning the direct relations of chemical composition of wet atmosphere deposits and that of drainage runoff obtained through simultaneous observations and measurements of these variables in the same sites (Bučienė *et al.* 1995; Šileika, Gaigalis 2007; Augustaitis *et al.* 2008). The objective of

this research is to reveal the relations between the concentrations of wet deposits and those of drainage runoff in a long time-scale (from the late 1980s till nowadays) using data of periodical and simultaneous measurements. In addition it was necessary to focus on the reasons which enabled the changes in different nutrient loads at various time-periods. In order to fulfil these tasks the data obtained in two research sites of the Middle Lithuania: Akademija and Graisupis, with agroecosystems dominated in the landscape have been analysed. The results of the analysis are presented and discussed below.

## DATA AND METHODS

The environmental monitoring in Lithuania started in 1967 (Girgždys 2000). Though the monitoring network was expanded, the chemical composition of wet deposits was measured only in four main sites: Žemaitija, Aukštaitija and Dzūkija National Parks (further NPs) and Preila atmosphere research station (further ARS) (Fig. 1). Currently the measurements in Dzūkija NP are cancelled.

In the Akademija research site, LIA, the matter migration experiment has been initiated in 1988 in order to study the different crop management systems; since then till 2000 the samples of wet precipitation for chemical analysis have been taken and analysed periodically (monthly or two time per month). The sampling has been fulfilled using precipitation receiver at 0.5 m height connected with a 3-litre bottle and fixed on the meteorological ground. Nitrate nitrogen is analysed potentiometrically, ammonium nitrogen and P are determined photometrically, and K is measured by flame photometry. Sulphate sulphur is analysed

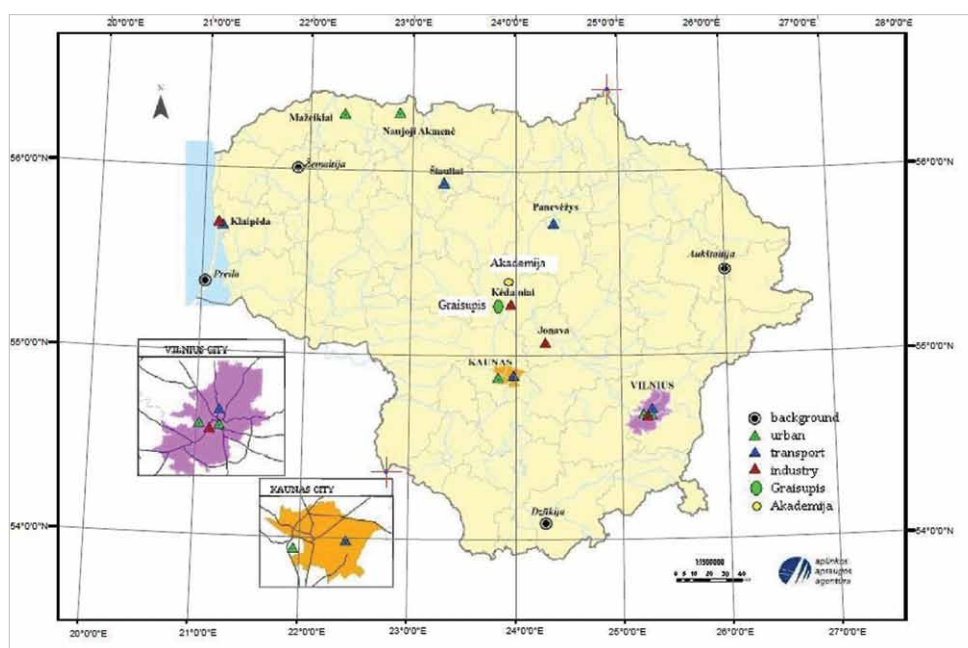
by turbidimetric method, calcium and magnesium is analysed trilonometrically, pH is measured potentiometrically. Data on quantity of wet deposits have been taken from Dotnuva Agrometeorological station located in Akademija research site.

The sampling of wet deposits started in 1997 for pH, nitrogen, phosphorus and other element analysis in the Graisupis agromonitoring site continues up to now. Nitrate is determined photometrically, total N is determined by its oxidation with peroxodisulphate to nitrate, followed by a photometry using phenol disulphoic acid. Phosphates are analysed photometrically using ammonium molybdate and total P by the same method after its oxidation using potassium peroxodisulphate. Potassium is determined by flame photometry, pH is measured potentiometrically, and ammonium nitrogen is determined spectrometrically. The same elements have been determined in the drainage runoff water samples periodically (BEAROP in Lithuania 1997).

In Scandinavian countries, like Sweden, the method of small drainage plots has been used for the research on comparison of different fertilization, crop and soil management treatments. The drainage system site is usually equipped with modern discharge measuring and sampling equipment, portable meteorological station (data-loggers, automatic samplers), that allows to make different research simultaneously. Drainage systems in Sweden are designed small – about 0.10–0.09 ha each (Gustafson 1988). The method of small drainage plots (1.4–5.9 ha) was first applied in Lithuania in 1963 to compare the water balance elements in the drained and not-drained soils (Bučienė *et al.* 1995). Later even smaller drainage plots were designed for the research purposes.

Physical-geographical conditions in Graisupis watershed, covering the area of 1415 ha, are typical for the Middle Lithuania. The watershed includes

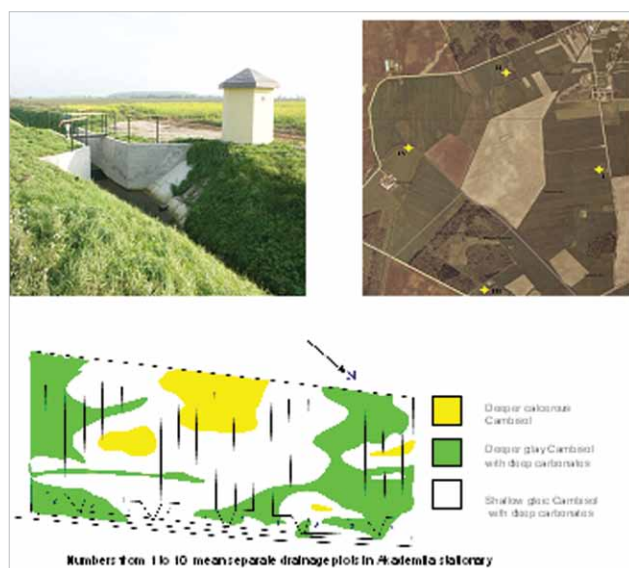
the Graisupis upstream drainage area. The Quaternary sediment layer consists of sandy loam and light loam material. The soil prevailing in the watershed as well as in the Demonstration farm is an Endocalcari-Endohypogleic Cambisol. The landscape is flat, 62–65 m above sea level. The main monitoring station is constructed about 1.3 km upstream of the outlet of the river Graisupis (Fig. 2). The Demonstration farm is a typical mixed farm with dairy cattle and cereal production. In this farm the monitoring of drainage runoff started in 1997 in the plot of 7.4 ha, that mainly consists



**Fig. 1** The national network of air quality monitoring in Lithuania and the agro-monitoring sites in the Middle Lithuania: Akademija and Graisupis. Adopted from <http://oras.gamta.lt/cms/index?rubricId=42123d1f-77ae-4fcd-bdcb-33dfcfc0500c>. LKS-94, © 2010, Nacionalinė žemės tarnyba prie Žemės ūkio ministerijos. Compiled by A. Bučienė, 2012.

of grass agroecosystems on both arable land and the pasture. A drainage runoff data logger, constructed at the outlet of the drainage system, has recorded leaching from the fields. For drainage discharge measurements, the V-notch ( $\alpha = 90^\circ$ ) Thomson weir is used, and water samples have been collected once a month for analysis. The analyses have been conducted at the laboratory of the Water Management Institute.

Crop management experiment in Akademija site has been performed also on Endocalcari-Endohypogleyic Cambisols consisting of sandy-loam and light loam in their texture (Fig. 2). The drainage plots in Akademija experimental site range in size from 0.34 to 0.38 ha each (32x107–119 m). Field drains are placed mainly at a depth around one metre in a 16 m distance on the average. Drainage water flow is registered continuously by a water-stage recorders. The site was previously



**Fig. 2** A. Graisupis stream monitoring post and landuse in the watershed (upper figure; photo by K. Gaigalis). B. Soil map of the drainage runoff measuring stationary in Akademija site (lower figure; after A. Bučienė *et al.* 2007).

managed as a long-term extensive grassland for milking cows (Bučienė *et al.* 2007). Drainage water for chemical analysis has been sampled every second week but more frequently during flood conditions. The nutrients and pH in drainage water samples have been determined by the same methods as in wet deposit samples. Analyses have been conducted at the analytical laboratory of Lithuanian Institute of Agriculture. The leached amount of different nutrients has been estimated by multiplying drainage

runoff accumulated during certain time period by the element concentration.

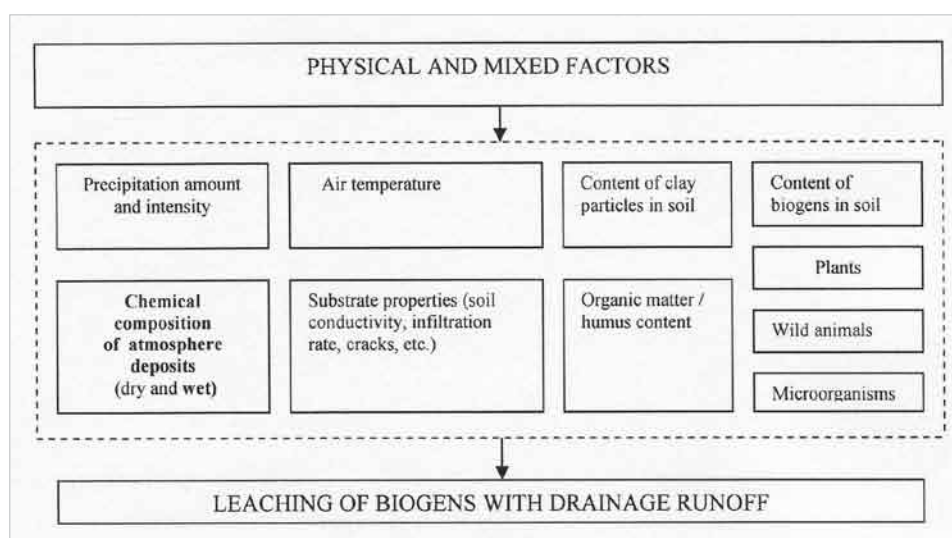
For the calculation of relations between the concentration of wet deposits and drainage runoff in Akademija site two treatments have been chosen: long-term pasture (LTP) and reference plot (REF) with four different crops: winter wheat, potatoes, barley with clover undersow, and first-year ley. During the second rotation potatoes have been changed by rape-seed. For regression-correlation analysis between different variables *STAT\_ENG for EXCEL vers. 1.55* have been used.

## RESULTS

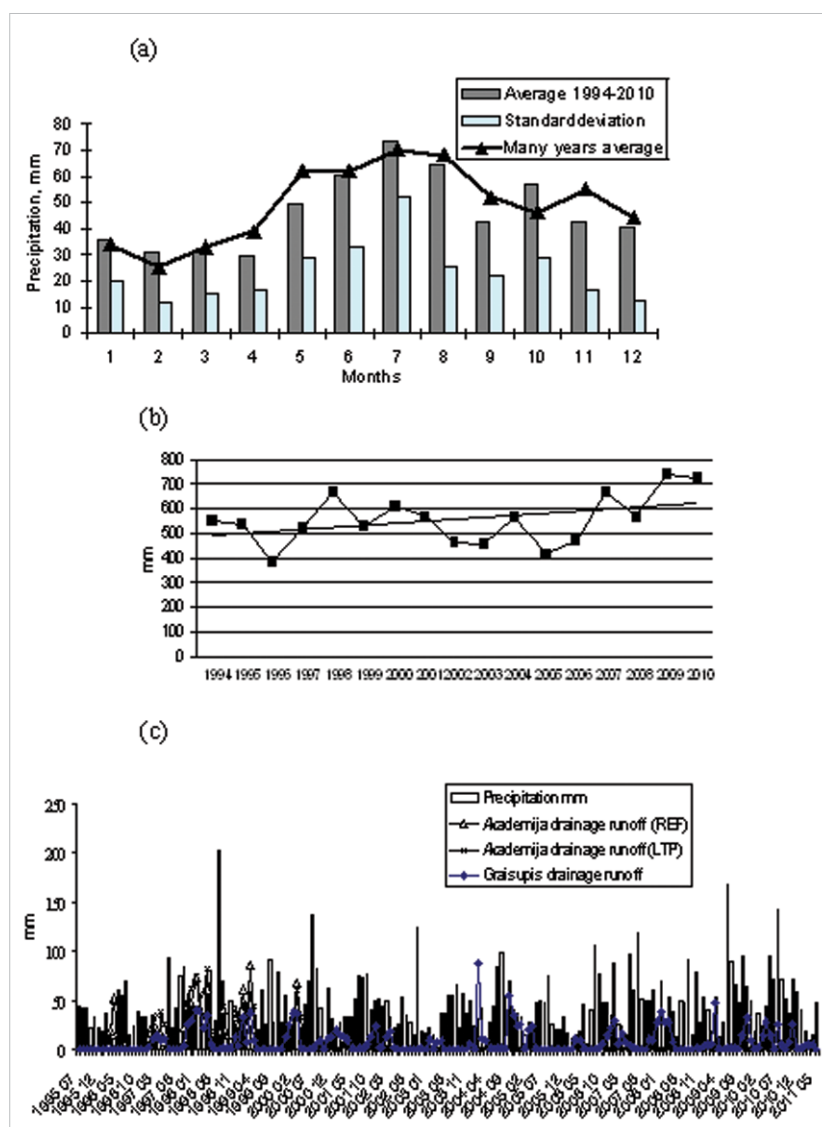
The factors, influencing the leaching of chemical elements with drainage runoff might be divided to the physical, anthropogenic and mixed. Two of these three are shown in Fig. 3. The precipitation rate is recognized as one of the most important physical factors, that effects on the nutrient leaching results, and the chemical composition of wet deposits is one of the mixed factors, that has been focused on in this research, particularly.

The long-term average precipitation in Dotnuva agrometeorological station is ranging in different months as it is shown in Fig. 4a. The average monthly rate from 1994 to 2010 was distributed close to it, except for October. The annual precipitation rate and the trend for 1994–2010 in mm is presented in Fig. 4b. Data show the slight increase trend which is a consequence of global climate warming in this latitude.

During the study period from 1994 to 2010 the meteorological situation was varying from the wettest hydrological years of 1997–1998, 1998–1999 and 2009–2010, to the driest ones – 1995–1996, 2002–2003, 2003–2004, 2005–2006 while the rest years were notable for medium values of precipitation and discharge rate. The data on monthly precipitation rate and drainage runoff values in REF, LTP treatments



**Fig. 3** Factors influencing the leaching of biogens with drainage runoff. Compiled by A. Bučienė, 2012.



**Fig. 4** Average monthly precipitation in mm in 1994–2010 plus standard deviation and many years average in Dotnuva agrometeorological station (a); annual precipitation rate and the trend for 1994–2010 in Dotnuva station (b); monthly values of precipitation rate and drainage runoff in mm in the REF and LTP treatments of Akademija site and Graisupis site in 1995–2011 (c). Compiled by A. Bučienė and K. Gaigalis, 2012.

(Akademija site) and Graisupis site (Fig. 4c) show, that drainage runoff in normal years makes about 20%, in wet years – close to 40% and in dry years about 7–10% of annual precipitation amount on the average.

The results of monthly chemical analysis of wet deposit samples have shown large deviation from the long-term annual average, and they have revealed some trends in time-scale in both research sites (Tables 1, 2). The Akademija site is found to contain the highest amounts of  $\text{SO}_4^{2-}\text{-S}$  and  $\text{NH}_4^+\text{-N}$  in wet deposits in 1988–1989. Later they were lower due to a development slowdown both in agriculture and industry. On the contrary, the contents of Ca and Mg in wet deposits are obtained, on the average, lower in 1988–1989 than in the following years. The highest contents of ammonium nitrogen in wet deposits at Graisupis site are determined in summer months, particularly in June, while the peak in phosphate phosphorus is in autumn months.

During 1988–1989 in Akademija site, the 11.5 kg ammonium nitrogen and 2.4 kg nitrate nitrogen, in total 13.9 kg of mineral N, as well as 0.3 kg  $\text{PO}_4\text{-P}$ , 4.5 kg K, 29 kg  $\text{SO}_4\text{-S}$ , 20 kg of Ca and 5 kg of Mg  $\text{ha}^{-1}$  is estimated to be deposited on the average per year (Fig. 5a). However, from 1990, because of economic slowdown the fuel consumption and industry decreased several times. The analysis from 1994 to 1999 has shown the decrease in nitrogen and sulphur, but slight increase in Ca and Mg deposits (Fig. 5b). During that period 7.6 kg of mineral nitrogen (1.9 kg nitrate nitrogen and 5.7 kg ammonium nitrogen), 0.35 kg  $\text{PO}_4\text{-P}$ , 4.3 kg K, 9.6

**Table 1** Average annual concentration of the main nutrients in wet deposits in  $\text{mg l}^{-1}$  in Akademija site.

Year	$\text{SO}_4^{2-}\text{-S}$	$\text{NO}_3^-\text{-N}$	$\text{NH}_4^+\text{-N}$	$\text{PO}_4^{3-}\text{-P}$	$\text{K}^+$	$\text{Ca}^{+2}$	$\text{Mg}^{+2}$
1988	5.54±1.05	0.83±0.14	2.30±0.58	0.070±0.010	1.00±0.19	4.16±0.66	1.45±0.24
1989	5.48±0.89	0.37±0.17	2.57±0.35	Nd	1.38±0.28	3.41±0.62	0.98±0.11
1994	1.42±0.28	0.10±0.01	0.84±0.27	0.040±0.028	0.63±0.14	8.31±1.91	1.13±0.25
1995	Nd	1.16±1.00	0.39±0.20	0.080±0.038	0.23±0.17	4.45±2.43	0.73±0.12
1996	3.05±0.62	0.25±0.17	0.82±0.09	0.030±0.006	1.11±0.24	4.44±1.23	1.51±0.50
1997	1.20±0.20	1.15±0.58	0.79±0.27	0.110±0.030	1.71±0.44	5.02±1.86	1.51±0.49
1998	1.84±0.15	0.03±0.02	0.47±0.18	0.054±0.022	0.29±0.12	8.22±6.44	4.57±4.01
1999	1.87±0.05	0.31±0.30	1.39±0.48	0.158±0.100	2.44±1.06	4.06±0.80	0.97±0.01

Nd – not determined.

**Table 2** Average annual concentration of the main nutrients in wet deposits in mg l<sup>-1</sup> and total number of animals in Demonstration farm of Graisupis site.

Year	NO <sub>3</sub> <sup>-</sup> -N	NH <sub>4</sub> <sup>+</sup> -N	PO <sub>4</sub> <sup>-3</sup> -P	K <sup>+</sup>	Total number of animals
1996	0.53±0.38	0.95±0.54	0.020±0.015	Nd	21
1997	1.26±1.17	1.39±1.14	0.128±0.096	Nd	41
1998	0.28±0.18	0.89±0.70	0.039±0.036	4.04±3.67	65
1999	1.02±0.58	2.24±1.43	Nd	5.56±3.85	64
2000	0.82±0.37	1.72±0.83	0.137±0.100	Nd	64
2001	0.84±0.44	1.43±0.54	0.027±0.021	Nd	85
2002	0.91±0.50	2.19±1.53	0.276±0.231	Nd	104
2003	0.83±0.74	2.39±1.43	0.156±0.140	Nd	153
2004	1.10±0.66	1.51±1.21	0.090±0.059	Nd	175
2005	0.94±0.70	2.85±1.86	0.062±0.049	Nd	204
2006	2.57±2.33	2.07±1.05	0.120±0.103	Nd	220
2007	1.02±0.69	2.09±1.07	0.058±0.017	Nd	270

kg SO<sub>4</sub>-S, 36.2 kg Ca and 7.0 kg ha<sup>-1</sup> Mg is deposited on the average per year.

In Graisupis site (Fig. 5a), the load of ammonium nitrogen is shown to increase from 5 to 12 kg ha<sup>-1</sup> from 1996 to 2007, the same for nitrate nitrogen is observed from 3 to 6 kg ha<sup>-1</sup>, and the sum of both nitrogen forms has increased from 9 to 12 kg ha<sup>-1</sup> on the average. The deposition of phosphate-phosphorus has not changed during that research period in Graisupis site. If to compare the results for 1996–1999 in both sites, they look quite relevant for the 1996–1998, but differ for the year 1999 concerning ammonium nitrogen load that is more than two time higher in Graisupis than in Akademija. Probably, the difference might be due to the increase in animal number in the Demonstration farm situated on the Graisupis watershed (Gaigalis *et al.* 2006) and because of impact of higher industrial emissions in Kėdainiai district with recovery of economics in further years. The increase in car number and transport intensity started already in 1994 in Lithuania that might have also influenced the total emissions of nitrogen in Kėdainiai district as well (Table 3).

In 1988–1989, the average pH of wet deposits in Akademija site is found to be about 6.6 (slightly acid). Since then it increases to 7.0 and more in 1994–1995, while in 1996 pH drops again to the 6.6 level (Fig. 5c). The second alkalisation period has lasted longer than 1999, even up to 2002, as data from Graisupis site demonstrate, but from 2003 pH of wet deposits becomes neutral, except the 2008 peak up to 7.5. Among the examined cations and anions in wet atmosphere deposits, the SO<sub>4</sub><sup>-2</sup>-S, Mg<sup>+2</sup> and Ca<sup>+2</sup> show a significant impact on the drainage runoff concentrations on Cambisols in Akademija site (Fig. 6, three upper graphs).

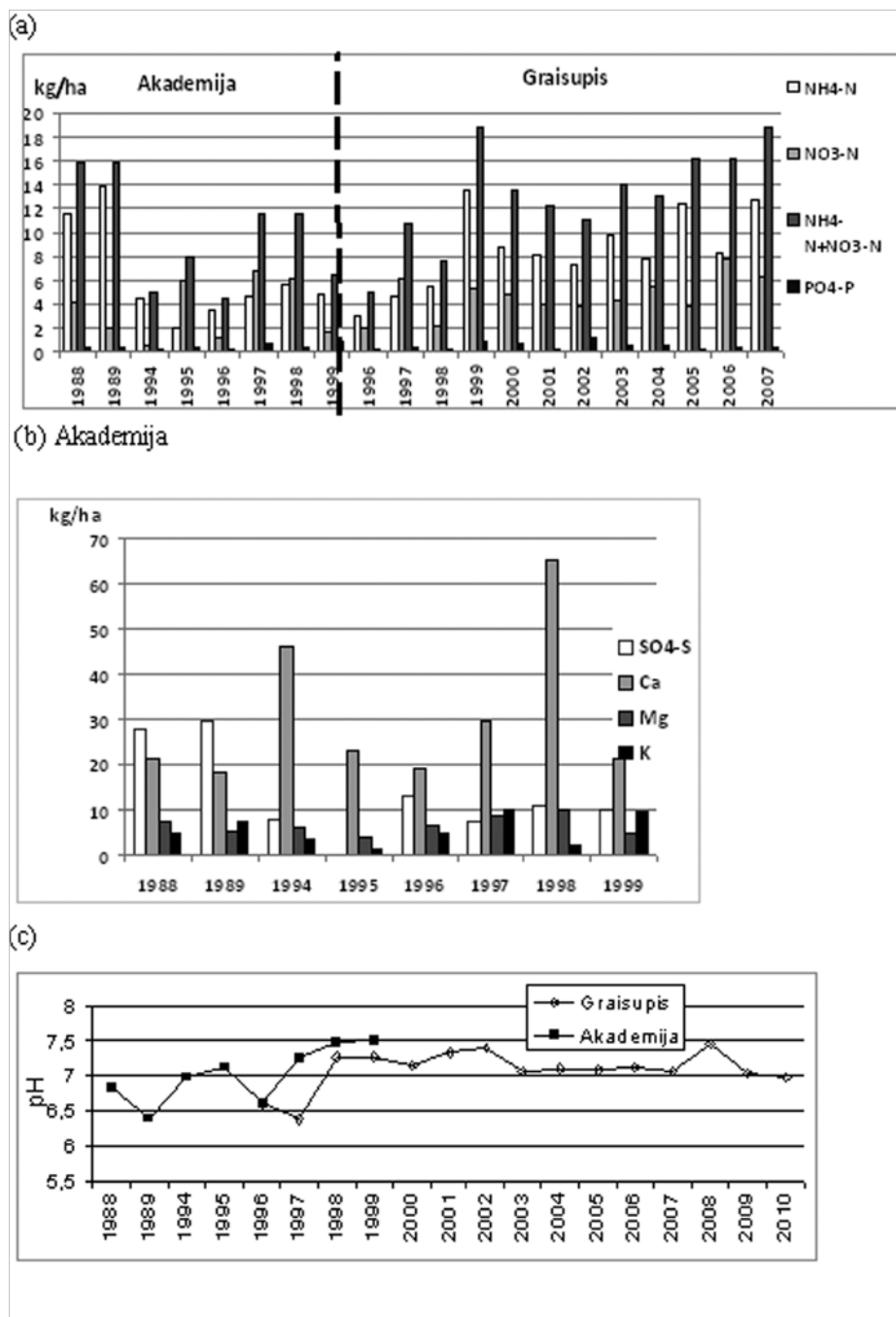
The increase in element contents in wet deposits is accompanied by that in drainage runoff in accordance to the exponential curve for SO<sub>4</sub>-S (R=0.59), and Ln(x) for Mg and Ca (R=0.61 and 0.35, consequently; R<sub>05</sub>=0.35) in REF treatment. But the correlation is found to be unreliable between the concentration of different nitrogen and phosphorus forms and potassium in wet deposits and drainage runoff (P>0.05) in the REF and LTP treatments. In Graisupis site, the contents of PO<sub>4</sub><sup>-3</sup>-P, NH<sub>4</sub><sup>+</sup>-N and NO<sub>3</sub><sup>-</sup>-N in drainage runoff are dependent on the contents of these substances in wet deposits (Fig. 6, three lower graphs): R=0.51, 0.49,

0.42, R<sub>05</sub>=0.42). Also there is a tendency observed in the correlation between concentration of K<sup>+</sup> in wet deposits and drainage runoff (R=0.34, P close to 0.05), but the data for K<sup>+</sup> are obtained for 1998–1999 only.

At the same time the negative correlation between the amount of wet atmosphere deposits (mm) and the concentration of NH<sub>4</sub><sup>+</sup>-N, mineral nitrogen and SO<sub>4</sub><sup>-2</sup>-S in mg l<sup>-1</sup> in Akademija site is estimated (Fig. 7 a, b, c). Other examined nutrients like Ca<sup>+2</sup>, Mg<sup>+2</sup>, K<sup>+</sup>, NO<sub>3</sub><sup>-</sup>-N and PO<sub>4</sub><sup>-3</sup>-P show the tendency of the same character (P>0.05). In Graisupis site, such a tendency is observed for phosphate phosphorus only (P > 0.05).

## DISCUSSION

According to the results obtained in Akademija site, sulphate sulphur load decreases from 29 to almost 10 kg ha<sup>-1</sup> year<sup>-1</sup> from 1988–1989 to 1994–1999 on the average. This decrease is mainly due to the drastic drop in sulphate content in wet deposits and that is in accordance to the research results obtained in different countries (Anderson, Downing 2006; Hertel *et al.* 2003; Schöpp *et al.* 2003; Kopáček *et al.* 1997; EMEP 2004; Matzner 1989) as well as in other places of Lithuania (Jasinevičienė, Šopauskienė 1999; Šopauskienė, Būdvytytė 1994, 1996). For example, sulphate contents in wet deposits decrease almost twice from 1994 to 1998 in the monitoring stations of NPs Aukštaitija, Žemaitija and Dzūkija, the later changes up to 2003 are not significant (Augustaitis *et al.* 2006). Sulphur and other ions in wet deposits have not been analysed after 1999 in Akademija. But according to the data obtained in other Lithuania towns and rural landscapes of that period (Jucevičienė 2003;



**Fig. 5** Average estimated loads of different forms of N and PO<sub>4</sub>-P with wet deposits in kg ha<sup>-1</sup> year<sup>-1</sup> in 1988–1999 in Akademija site and in 1996–2007 in Graisupis site (a); loads of SO<sub>4</sub>-S, Ca, Mg and K in kg ha<sup>-1</sup> year<sup>-1</sup> in 1988–1999 in Akademija site (b); the pH of wet deposits in Akademija and Graisupis sites (c). Compiled by A. Bučienė and K. Gaigalis, 2012.

Baužienė 2005) it is evident that there are no changes in SO<sub>4</sub>-S contents in 2000–2003 as compared to the 1995–1999 period. Consequently, the sulphur content in wet deposits remains at the same level. Similar trends have been observed in Germany, Poland and Latvia (EMEP 2004).

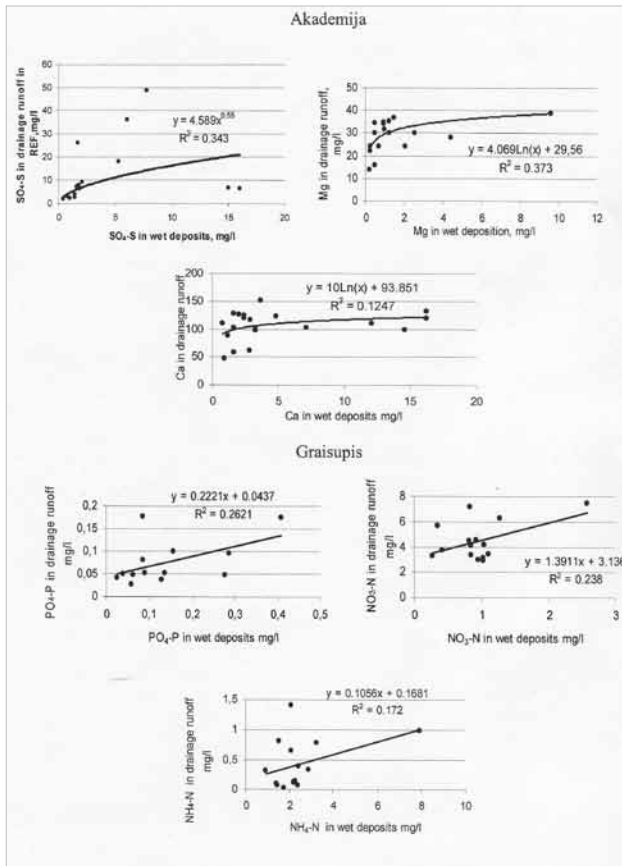
The research fulfilled in Žemaitija, Dzūkija and Aukštaitija NPs as well as Preila ARS has shown the prevalence of alkalinisation of precipitation in 1994–1998 in Aukštaitija and Dzūkija NPs territory,

acidification in 1996–1998 in Žemaitija NP area, and acidification in all three NPs during 1999–2001–2003 (Samuila 1999; Šopauskienė *et al.* 2006; Jasinevičienė, Šopauskienė 2006). However, no acidification of wet deposits is observed for that period in Akademija, but the acidification is determined in 1988–1989 and in 1995–1996 instead. The alkalinisation in 1997–2002 in both Akademija and Graisupis sites resembles that in the areas of Aukštaitija and Dzūkija NPs, just with a certain delay.

Concerning the mineral nitrogen concentrations and load, the tendency of its increase in wet deposits has been determined in the Middle Lithuania mainly due to the increase in ammonium nitrogen concentration from 1999 to 2007. This might be stipulated by the local reasons: increase in animal number in Demonstration farm on the Graisupis watershed and also because of higher traffic load and industrial emissions in Kėdainiai district with the recovery of economy. The research in NPs and Preila ARS does not show such a tendency (Šopauskienė, Jasinevičienė 2006), thus it could be the local phenomena.

In general, the precipitation amount in the Middle Lithuania according to the data of Dotnuva station shows a slight trend of increase from

1994 to 2010, and it is in accordance to other investigations conducted in the Eastern Lithuania, Aukštaitija NP, from 1994 to 2004 (Baužienė 2005). All this is thought to be a consequence of global climate warming. However at the same time, a tendency of decrease in precipitation rate during the vegetation period, and its increase in the cold season is observed (Pauliukevičius 2004; Rimkus *et al.* 2009). From the ecosystem point of view this is more threatening because the plants and



**Fig. 6** Dependence of  $\text{SO}_4^{2-}\text{-S}$ ,  $\text{Mg}^{+2}$ ,  $\text{Ca}^{+2}$ ,  $\text{PO}_4^{3-}\text{-P}$ ,  $\text{NO}_3\text{-N}$ ,  $\text{NH}_4\text{-N}$  concentration in drainage runoff in  $\text{mg l}^{-1}$  on the concentration of those substances in wet deposits in  $\text{mg l}^{-1}$  in Akademija and Graisupis sites. Compiled by A. Bučienė and K. Gaigalis, 2012.

crops use a little water and nutrients from soil during cold season, thus they can easily be leached with higher precipitation and drainage runoff amount. The nitrogen is one of nutrients most vulnerable for leaching during the cold season.

**Table 3** Total number of road vehicles and individual cars in Lithuania in 2000–2009 and total emission in tons from stationary sources in Kėdainiai district.

Year	Total number of road vehicles	Of that total number of cars	Total emission from stationary sources in Kėdainiai district, t
2000	1 286 392	1 065 415	No data available
2001	1 383 724	1 116 473	1990
2002	1 479 099	1 180 718	No data available
2003	1 580 476	1 260 034	No data available
2004	1 634 354	1 318 562	3000
2005	1 789 657	1 457 954	No data available
2006	1 950 302	1 595 928	2895
2007	1 973 622	1 596 186	3416
2008	2 106 848	1 704 063	3149
2009	2 133 720	1 726 462	2767*

\* For the beginning of year 2010. Web source: <http://www.stat.gov.lt/lt/pages/view/?id=2463>

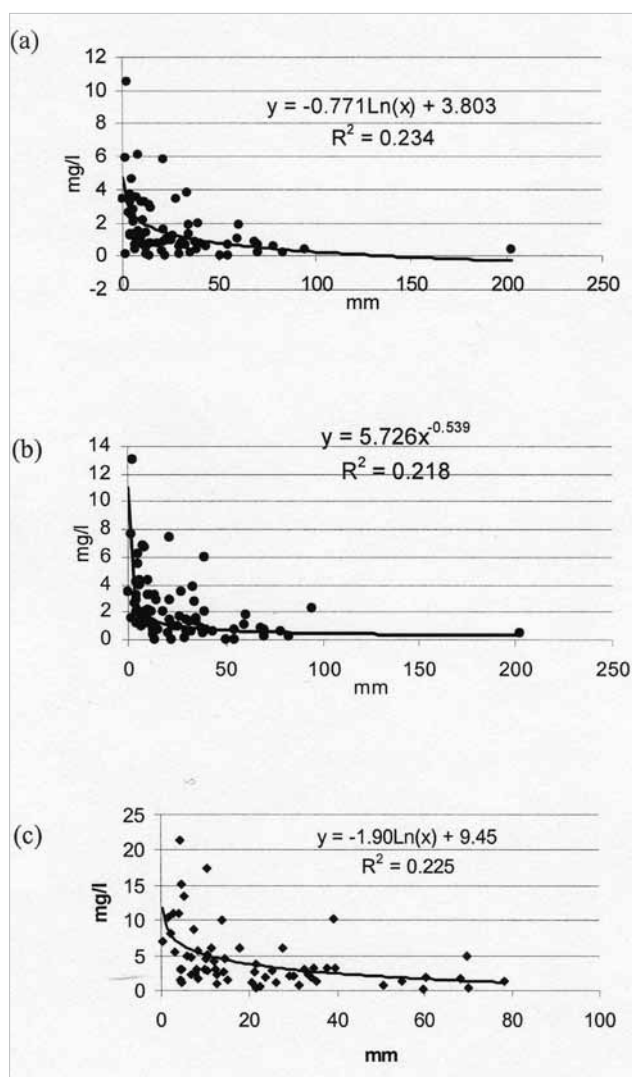
The content of nutrients  $\text{SO}_4^{2-}\text{-S}$ ,  $\text{Mg}^{+2}$ ,  $\text{Ca}^{+2}$ ,  $\text{NO}_3\text{-N}$ ,  $\text{NH}_4\text{-N}$  and  $\text{PO}_4^{3-}\text{-P}$  in wet deposits positively affected that in drainage runoff on the Cambisols of Middle Lithuania. A non-linear user-specified regression applied for the yearly observation data in three agricultural watersheds in the western (Lyžena), middle (Graisupis) and south-eastern (Vardas) parts of Lithuania has revealed, that the precipitation plays the key role in N contents in these streams ( $R^2=0.72$  and  $R^2=0.61$ ). The N content in the Lyžena stream depends mostly on N content in precipitation and weather temperature ( $R^2=0.58$  and  $R^2=0.54$ ) (Šileika, Gaigalis 2007).

The negative correlation between the amount of wet atmosphere deposits (mm) and the concentration of ammonium nitrogen, mineral nitrogen and  $\text{SO}_4\text{-S}$  in  $\text{mg l}^{-1}$  has been determined in the Akademija site. This can be explained by the fact, that with higher amount of precipitation, the air mass is more washed up and thus contains less ions than that washed by the lower precipitation amount (Kvietkus *et al.* 2011). The same trends have been determined in the NPs and Preila ARS (Šopauskienė, Jasinevičienė 2006).

Still there are many unsolved issues with phosphorus load with deposits. Some authors refer that dry deposition could be more important than wet, because P deposition originates from soil and does not become incorporated into rainfall to a great degree (Anderson, Downing 2006). And opposite, it is suggested (Peters, Reese 1995) that P in dry deposition may be quite readily available to biota (such as algae and Cyanobacteria) compared to wet deposition, as there is a greater percentage of soluble reactive phosphorus in dry deposition. Thus, the analysis of both P in wet and dry deposition should receive more emphasis in a future research.

## CONCLUSIONS

The trend of increase in annual precipitation rate as a consequence of the climate warming has been determined from 1994 to 2010 in the middle of Lithuania according to the data of Dotnuva agrometeorological station. The acidification of wet deposits is observed in 1988–1989, and in 1995–1996, and alkalisation is seen in 1997–2002 and 2007–2008. The decrease in sulphate sulphur load from 29 to almost 10  $\text{kg ha}^{-1} \text{ year}^{-1}$  and mineral nitrogen from 13.9 to 7.6  $\text{kg ha}^{-1} \text{ year}^{-1}$ , also slight increase in calcium and magnesium has been



**Fig. 7** Correlation between amount of wet atmosphere deposits (mm) and the concentration of ammonium nitrogen (a), mineral nitrogen (b), and  $\text{SO}_4^{2-}\text{-S}$  in  $\text{mg l}^{-1}$  (c), Akademija site. Compiled by A. Bučienė, 2012.

taking place from 1988–1989 to 1994–1999 due to the economy reduction: decrease in transport load, fuel consumption and industrial emission.

The tendency of increase of both ammonium and nitrate nitrogen concentration and load of these nutrients in wet deposits has been observed from 1999 to 2007. This could be related to the increase in animals number in the Demonstration farm and also to the impact of higher industrial emission due to economy recovery. The road traffic began to increase significantly from 1994 because of a rise in vehicles number.

The phosphate phosphorus and potassium concentration and load with wet deposits does not change during the period studied. The contents of  $\text{SO}_4^{2-}\text{-S}$ ,  $\text{Mg}^{2+}$ ,  $\text{Ca}^{2+}$ ,  $\text{NO}_3^{-}\text{-N}$ ,  $\text{NH}_4^{+}\text{-N}$  and  $\text{PO}_4^{3-}\text{-P}$  in wet deposits positively affected those in the drainage runoff on the Cambisols of the Middle Lithuania. The negative correlation between the amount of wet atmosphere deposits (mm) and the concentration of  $\text{NH}_4^{+}\text{-N}$ , mineral

nitrogen and  $\text{SO}_4^{2-}\text{-S}$  in  $\text{mg l}^{-1}$  has been determined in the Akademija stationary site.

In a future research, more attention is to be given to both the chemical composition and amount of wet and dry deposition and its impact on chemical composition of drainage runoff. Different forms of P should be analysed in wet and dry atmosphere deposits in the first order, because this nutrient is among the most responsible elements in water eutrophication.

## Acknowledgements

Authors are thankful to the reviewers Dr. Barbro Ulén (Uppsala) and Professor Arūnas Bukantis (Vilnius) for valuable remarks and suggestions to improve the quality of this paper. The staff of laboratories of Lithuanian Institute of Agriculture and Water Management Institute is acknowledged for their helpful assistance.

## References

- Anderson, K. A., Downing, J. A., 2006. Dry and wet atmospheric deposition of nitrogen, phosphorus and silicon in an agricultural region. *Water, Air and Soil Pollution* 176, 351–. <http://dx.doi.org/10.1007/s11270-006-9172-4>
- Augustaitis, A., Arbačiauskas, K., Baužienė, I., Eitminavičiūtė, I., Girgždienė, R., Kliučius, A., Mažeikytė, R., Mozgeris, G., Rašomavičius, V., Šopauskienė, D., 2006. *Sąlygiškai natūralių ekosistemų kompleksiškas monitoringas. Dešimtmečio rezultatai*. [Complex monitoring of relatively natural ecosystems. The decade results], , Lututė, 111 pp. [In Lithuanian].
- Augustaitis, A., Augustaitienė, I., Kliučius, A., Pivoras, G., Bendoravičius, B., Šopauskienė, D., Jasinevičienė, D., Baužienė, I., Eitminavičiūtė, I., Arbačiauskas, K., Mažeikytė, R., 2008. N deposition, balance and benefit in the forest ecosystem of main landscape types of Lithuania. *International Journal of Environmental Studies* 65 (21), No. 3, 339–359.
- Baužienė, I., 2005. Flux of sulphur in the geosystem of Aukštaitija complex monitoring station. *Annales Geographicae* 38 (1), 72–79. [In Lithuanian with English summary].
- BEAROP in Lithuania, 1997. A project within the Baltic Agricultural Run-off Programme BAAP, Final report 1994–1997. Lithuanian team: Šileika, A.-S., Gaigalis, K., Kutra, G., Bučienė, A., Strusevičius, Z.; Swedish team: Göran Carlson, Staffan Steineck, Arne Gustafson, Stefan Lofgren, Christine Jakobsson, Alf Gustafson. Lithuanian Institute of Water Management, Swedish Institute of Agricultural Engineering, Kėdainiai, Uppsala, 1997, 128 pp.
- Bučienė, A., Antanaitis, Š., Mašauskienė, A., Šimanskaitė, D., 2007. Nutrients N, P losses with drainage runoff and field balance as a result of crop management. *Communications in Soil Science and Plant Analysis* 38 (15 & 16), 2177–2195. <http://dx.doi.org/10.1080/00103620701549124>



- Bučienė, A., Mašauskas, V., Juozapaitis, A., 1995. Application of small drainage system method in the matter migration and balance research. *Proceedings of the Fourth Regional Conference on Mechanization of Field Experiments (IAMFE/BALTIC '95)*, Kaunas/Dotnuva, Lithuania, 93–99.
- Bučienė, A., Šileika, A. S., Gaigalis, K., 1998. Nutrient balance and management in r. Graisupis watershed and demonstration farm. *Monographs in System Ecology 2*, Klaipėda University, Klaipėda, 6–11.
- Deelstra, J., Kverno, S. H., Granlund, K., Šileika, A. S., Gaigalis, K., Kyllmar, K., Vagstad, N., 2009. Runoff and nutrient losses during winter periods in cold climates – requirements to nutrient simulation models. *Journal of Environmental Monitoring 11*, 602–609. <http://dx.doi.org/10.1039/b900769p>
- Djordjic, F., Börling, K., Bergström, L., 2004. Phosphorus leaching in relation to soil type and soil phosphorus content. *Journal Environmental Quality 33*, 678–684. <http://dx.doi.org/10.2134/jeq2004.0678>
- EMEP, 2004. EMEP Assessment, Part II, National Contributions, ed. by J. Bartnicki and G. Lovbländ, Oslo. [Interactive: <http://www.emep.int> ; 2012–06-05].
- Fagerberg, B., Salomon, E., Steineck, S., 1993. *The computer program NPK-FLO*. Uppsala, 47 pp.
- Gaigalis, K., Kutra, G., Šmitienė, A., 2007. Nitrogen load assessment and pollution mitigation measures in the Dovinė watershed. *Ekologija 53 (2)*, 37–43.
- Gaigalis, K., Šileika, A. S., Šmitienė, A., 2006. Changes of nitrogen and phosphorus concentrations in streams of agricultural catchments. *Water Management Engineering 30 (50)*, 44–56. [In Lithuanian with English summary].
- Girgždys, A., 2000. *Aplinkos monitoringas* [Environmental monitoring]. Vilnius, Technika, 120 pp. [In Lithuanian].
- Gustafson, A., 1988. Simulation of nitrate leaching from arable land in Southern Sweden. *Acta Agriculturae Scandinavica 38 (1)*, 13–23. <http://dx.doi.org/10.1080/00015128809436584>
- Gužys, S., Arlauskienė, E.A., 2001. Relation between agricultural intensity and productivity of agroecosystem and matter migration. *Water Management Engineering 16 (38)*, 94–110. [In Lithuanian with English summary].
- Gužys, S., 2002. Ecological estimation of cropping intensity. *Ekologija 4*, 39–48. [In Lithuanian with English summary].
- HELCOM 2002 : *Fourth periodic assessment of the state of the marine environment of the Baltic Sea area*, 1994–1998. Background Document, Baltic Sea Environment Proceedings No. 82 B, 216 pp.
- Hertel, O., Ambelas Skjøth, C., Brandt, J., Christensen, J. H., Frohn, L. M., Frydendall, J., 2003. Operational mapping of atmospheric nitrogen deposition to the Baltic Sea. *Atmospheric Chemistry and Physics 3*, 2083–2099. [Interactive: <http://www.atmos-chem-phys.org/acp/3/2083> ; 2012–06-10]. <http://dx.doi.org/10.5194/acp-3-2083-2003>
- Jasinevičienė, D., Šopauskienė D., 1999. Changes of sulphate concentration in atmosphere deposits in Lithuania. *Environmental Engineering 4*, 187–193. [In Lithuanian with English summary].
- Jucevičienė, N., 2003. The research on pollution of precipitation in Kaunas city. *Environmental Research, Engineering and Management 2 (24)*, 44–47. [In Lithuanian with English summary].
- Kopáček, J., Prochazkova, L., Hejzlar, J., 1997. Trends and seasonal patterns of bulk deposition of nutrients in Czech Republic. *Atmospheric Environment 31*, 797–808. [http://dx.doi.org/10.1016/S1352-2310\(96\)00261-0](http://dx.doi.org/10.1016/S1352-2310(96)00261-0)
- Kvietkus, K., Šakalys, J., Valiulis, D., 2011. Trends of atmospheric heavy metal deposition in Lithuania. *Lithuanian Journal of Physics 51 (4)*, 359–369. <http://dx.doi.org/10.3952/lithjphys.51413>
- Lindfors, V., Joffre, S., Damski, J., 1993. Meteorological variability of the wet and dry deposition of sulphur and nitrogen compounds over the Baltic Sea. *Water, Air and Soil Pollution 66*, 1–28. <http://dx.doi.org/10.1007/BF00477058>
- Mašauskas, V., Antanaitis, Š., Lazauskas, S., 2006. Content of nitrates in drainage and ground water from grassland and arable land. In Present-day problems of soil fertility increase and their protection from degradation 27–29 June 2006, Minsk, 161–162.
- Matzner, E., 1989. Acidic precipitation: case study: Solling, West Germany. In D. C. Adriano and M. Havas (eds), *Advances in environmental science: acid precipitation, Vol. 1*, Berlin Heidelberg New York, 39–83. [http://dx.doi.org/10.1007/978-1-4612-3616-0\\_2](http://dx.doi.org/10.1007/978-1-4612-3616-0_2)
- Peters, N. E., Reese, R. S., 1995. Variations of weekly atmospheric deposition for multiple collectors at a site on the shore of Lake Okeechobee, Florida. *Atmospheric Environment 29*, 179–. [http://dx.doi.org/10.1016/1352-2310\(94\)00233-B](http://dx.doi.org/10.1016/1352-2310(94)00233-B)
- Pauliukevičius, H., 2004. Specific features of small river runoff in years with dry warm season. *Annales Geographicae 37 (1-2)*, 18–26. [In Lithuanian with English summary].
- Povilaitis, A., 2004. Phosphorus trends in Lithuanian rivers affected by agricultural non-point pollution. *Environmental Research, Engineering and Management 4 (30)*, 17–27.
- Povilaitis, A., 2006. Impact of agriculture decline on nitrogen and phosphorus loads in Lithuanian rivers. *Ekologija 1*, 32–39.
- Rosenberg, R., Elmgren, R., Fleischer, S., Jonsson, P., Persson, G., Dahlin, H., 1990. Marine eutrophication case studies in Sweden. *Ambio 19*, 102–108.
- Rimkus, E., Kažys, J., Bukantis, A., 2009. Predictions of heavy precipitation in the 21st century in Lithuania, based on the CCLM model. *Geografija 45 (2)*, 122–130. [In Lithuanian with English summary].
- Samuila, M., 1999. Simulation of some hydrological indices in the Aukštaitija integrated monitoring territory. *Annales Geographicae 32*, 260–277.
- Schöpp, W., Posch, M., Mylona, S., Johansson, M., . Long-term development of acid deposition (1880–2030) in sensitive freshwater regions in Europe. *Hydrology*

- and *Earth System Science* 7, 436–446. <http://dx.doi.org/10.5194/hess-7-436-2003>
- Šileika, A. S., Gaigalis, K., 2007. Influence of physical and anthropogenic factors on nitrogen leaching in small catchments. *Water Management Engineering* 31 (51), 36–46. [In Lithuanian with English summary].
- Šileika, A. S., Gaigalis, K., Kutra, G., Šmitienė, A., 2005. Factors affecting N and P losses from small catchments (Lithuania). *Environmental Monitoring and Assessment* 102 (3), 359–374. <http://dx.doi.org/10.1007/s10661-005-6033-3>
- Šmitienė, A., Larson, M., Gaigalis, K., Baigys, G., 2005. Calibration and verification of drain water discharge by the SOILNDB model under Lithuanian conditions. *Water Management Engineering* 2 (5), 25–32.
- Šopauskienė, D., Būdvytytė, D., 1996. Atmospheric precipitation and throughfall chemistry in coniferous forests of Lithuania. *Atmospheric Physics* 18 (1), 5–.
- Šopauskienė, D., Būdvytytė, D., 1994. Chemical characterization of atmospheric aerosol in rural site in Lithuania. *Atmospheric Environment* 28, 1291–. [http://dx.doi.org/10.1016/1352-2310\(94\)90275-5](http://dx.doi.org/10.1016/1352-2310(94)90275-5)
- Šopauskienė D., Jasinevičienė D., 2006. Changes in precipitation chemistry in Lithuania for 1981-2004. *Journal of Environmental Monitoring* 8 (3), 347–352. <http://dx.doi.org/10.1039/b516877e>
- Tumas, R., 1997. Farming and the quality of stream water. *Environmental Engineering* 2 (8), 25–. [In Lithuanian with English summary].
- Tumas, R., 1999. Hydroecological evaluation of small Lithuanian rivers. In Reduction of agricultural runoff to the Baltic Sea, *Proceedings of International Conference 8-9 September 1999*, 29–33.
- Ulevičius, V., Byčenkienė, S., Senuta, K., 2009. Critical loads of sulphur and nitrogen for terrestrial ecosystems in Lithuania. *Lithuanian Journal of Physics* 49 (2), –. <http://dx.doi.org/10.3952/lithjphys.49201>
- Wright, R. F., Larssen, T., Camarero, L., Cosby, B. J., Ferrier, R. C., Helliwell, R., Forsius, M., Jenkins, A., Kopáček, J., Majer, V., Moldan, F., Posch, M., Rogora, M., Schöpp, W., 2005. Recovery of acidified European surface waters. *Environmental Science and Technology* 39 (3), 64A–72A. <http://dx.doi.org/10.1021/es0531778>