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Deposition of Acidifying and Eutrophying Pollutants in Southern Switzerland from 1988 to 2013

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Riassunto: Gli ossidi di zolfo e azoto emessi durante i processi di combustione e l'ammoniaca prodotta prevalentemente dall'agricoltura possono essere trasportati per lunghe distanze, trasformati e poi depositati causando eutrofizzazione e acidificazione degli ecosistemi sensibili. A causa della sua vicinanza con la Pianura Padana ricca di emissioni e delle sue precipitazioni generalmente abbondanti, il Sud della Svizzera è particolarmente esposto alla deposizione di inquinanti antropici.
Per il Canton Ticino sono state calcolate le mappe delle deposizioni totali di zolfo, azoto e acidità potenziale per i cinque periodi 1988-1992, 1993-1997, 1998-2002, 2003-2007, 2008-2012, sommando le mappe delle deposizioni umide con quelle delle deposizioni secche. Le prime sono state ottenute moltiplicando le mappe delle precipitazioni con quelle delle concentrazioni nelle piogge, queste ultime calcolate con regressioni lineari multiple descriventi le concentrazioni in funzione dei parametri latitudine, longitudine e altitudine. Le mappe delle deposizioni secche sono state calcolate da Meteotest.
I risultati mostrano come dall'inizio del periodo di monitoraggio le deposizioni di zolfo in particolare, ma anche quelle di azoto, sono diminuite significativamente: le prime grazie alla diminuzione del contenuto di zolfo negli olii da riscaldamento e alla sostituzione del carbone ricco di zolfo con altri combustibili fossili, le seconde principalmente per via dell'introduzione dei catalizzatori delle automobili e dei sistemi DeNOx negli impianti di combustione stazionari. A causa della diminuzione delle deposizioni di zolfo e azoto sono anche diminuite in modo significativo le deposizioni di acidità potenziale.
Il confronto delle mappe delle deposizioni con i carichi critici di azoto e di acidità degli ecosistemi più sensibili ha mostrato che, se da un lato le deposizioni di acidità sono attualmente scese nella maggior parte dei casi al di sotto dei carichi critici, dall'altro le deposizioni di azoto superano tuttora i carichi critici. Ciò significa che le emissioni di azoto devono essere ulteriormente diminuite.

Parole chiave: deposizione atmosferica, analisi dei trend, carichi critici, acidificazione, azoto, zolfo

Abstract: Sulphur and nitrogen oxides from combustion processes and ammonia from agriculture can be transported over long distances, transformed and then deposited, causing acidification and eutrophication of sensitive ecosystems. Because of its proximity to the emission rich Po Plain and of its generally abundant precipitations, Southern Switzerland is particularly exposed to deposition of anthropogenic pollutants.
Total sulphur, nitrogen and potential acid deposition maps for the Canton of Ticino for the 5 time periods 1988-1992, 1993-1997, 1998-2002, 2003-2007, 2008-2012 were calculated by adding up wet with dry deposition maps. Wet deposition maps were obtained by multiplying precipitation maps with rainwater concentration maps, calculated with multiple linear regression equations describing rainwater concentrations as a function of latitude, longitude and altitude. Dry deposition maps were delivered by Meteotest.
The results show that, since the beginning of the monitoring period, deposition of sulphur decreased substantially and, to a smaller extent, also nitrogen: the first due to the decrease of the sulphur content in liquid fuels and the partial substitution of sulphur rich coal with other fossil fuels, the second mainly because of the equipment of cars with catalytic converters and stationary combustion sources with DeNOx-systems. As a consequence of reduced sulphur and nitrogen deposition, deposition of potential acidity also decreased significantly.
The comparison of the calculated deposition maps with the critical loads of nitrogen and acidity of the most sensitive ecosystems revealed that at present depositions of acidity fall mostly below the critical limits, whereas depositions of nitrogen still exceed critical loads, meaning that further emission reductions of reactive nitrogen compounds are necessary.

Keywords: atmospheric deposition, trend analysis, critical loads, acidification, nitrogen, sulphur

INTRODUCTION

After emission into the atmosphere, sulphur and nitrogen oxides from combustion processes and ammonia from agriculture can be transported over long distances, transformed and then loaded on natural ecosystems as wet and dry deposition. Consequences are acidification and eutrophication of the most sensitive ecosystems like forests, alpine and subalpine grasslands, fens, bogs, heathlands and nutrient poor or acid-sensitive surface waters. For these ecosystems, in the framework of the Convention on Long-range Transboundary Air Pollution (CLRTAP) of the United Nations Economic Commission for Europe (UNECE), critical loads of nitrogen and acidity, below which no significant harmful effects on the chosen environments are expected to occur, have been calculated.

Switzerland regularly submits its emission inventories to the CLRTAP (Heldstab et al., 2014). Emissions of sulphur and nitrogen oxides started to increase steeply after the Second World War. Sulphur dioxide (SO_2) reached its maximum between 1965 and 1980, while nitrogen oxides (NO_x) peaked around 1985. From 1980 to 2012 the decrease of anthropogenic sulphur and nitrogen oxides emissions is as follows: SO_2 by 90% from 112 kt to 11 kt and NO_x (expressed as NO_2) by 59% from 168 kt to 69 kt. The reduction in emissions of anthropogenic ammonia (NH_3) was much lower: between 1980 and 2012, they decreased only by 25% from 82 kt to 62 kt. The reduction of SO_2 emissions has been mainly caused by a reduction of the sulphur content in heating oils and the partial substitution of sulphur rich coal with other fossil fuels, while the decrease of NO_x emissions has been mainly determined by the equipment of cars with catalytic converters and stationary combustion sources with DeNOx-systems (catalytic denitrificators that transform NO_x into N_2 and water after injection of anhydrous ammonia, aqueous ammonia or urea). The decrease in ammonia emissions can be explained to a large extent by a reduction of the number of animals in agriculture and to changes in production techniques in favor of animal well-being.

Because of its particular topography and meteorology, the air quality in Southern Switzerland is not only influenced by local emissions but also by transboundary air pollution originating from the Po Plain in Italy and particularly from the heavily polluted urban area of Milan. Wet deposition gets highly influeced by this phenomena. In fact, wet deposition in Southern Switzerland is mainly determined by warm, humid air masses originating from the Mediterranean Sea, passing over the Po Plain and colliding with the Alps. In Italy emissions decreased similarly to Switzerland. Since 1990, SO_2, NO_x and NH_3 decreased by 90%, 58% and 14%, respectively (Romano et al., 2014)*.

Thanks to the decrease in emissions, air quality also improved. In the Canton of Ticino since 1990 annual mean concentrations of SO_2 and NO_2 at Lugano decreased by 94% and 40%, respectively (UACER, 2013).

But what about deposition of sulphur and nitrogen? Did they also decrease and how much? And what about the critical loads for sensitive ecosystems, are they still exceeded? This paper aims to answer to these questions for Southern Switzerland, where monitoring of wet deposition begun already in the 1980's by the Section of air, water and soil protection of the Department of the territory of the Canton of Ticino. Since the late 1990's, wet deposition data are monitored with the financial support of the Federal Office for the Environment.

* Note that the percentage reduction in emissions in Italy is comparable to that in Switzerland, but the reduction in Switzerland started earlier (1980 for SO_2 and 1985 for NO_x).

METHODS

Wet deposition has been sampled weekly with wet-only samplers at 9 different sites in the Canton of Ticino: Acquarossa (since 1990), Monte Brè (since 1995), Bignasco (since 2001), Locarno-Monti (since 1988), Lugano (since 1989), Robiei (since 1996), Sonogno (since 2001), Stabio (since 1990). Sampling sites were chosen along a south-north axis and at various altitudes (200-1900 m a.s.l.). Rain samples were analyzed for pH, alkalinity, conductivity and the main cations and anions. Geographic and chemical parameters, analytical methods and quantification limits are described in Steingruber & Colombo (2010) and Steingruber (2015). Monthly, yearly and 5-years mean concentrations were calculated by weighting weekly concentrations with the sampled precipitation volume, while monthly and yearly wet deposition were calculated by multiplying monthly and yearly mean concentrations with the precipitation volume measured at a meteorological sampling station located close to the sampling site. This procedure has been chosen in order to avoid underestimation of monthly and yearly depositions due to occasionally missing weekly samples.

Trend analysis of monthly mean concentrations and monthly wet depositions were performed with the seasonal Mann-Kendall test (Hirsch et al., 1982) with a correction among blocks of data (Hirsch & Slack, 1984). The two sided tests for the null hypothesis that no trend is present were rejected for p-values below 0.05. Estimates for temporal variations in rainwater quality were quantified with the seasonal Kendall slope estimator (Gilbert, 1987). All trend analysis were calculated with the CRAN package "rkt 1.3" (Marchetto, 2014).

Deposition maps of total sulphur and nitrogen, total oxidized and reduced nitrogen and potential acidity were calculated for the 5-years periods 1988-1992, 1993-1997, 1998-2002, 2003-2007, 2008-2012. They were obtained by adding up dry with wet deposition maps. Dry deposition maps of the gaseous compounds NH_3, NO_2, SO_2, HNO_3 and of the NH_4^+- and NO_3^--containing aerosols were provided by Meteotest. For base cations, values modelled by the European Monitoring and Evaluation Programme (EMEP) for the year 2000 were used for the calculations. Wet and dry deposition values of calcium, magnesium and potassium of the 3 main 50 x 50

km grids falling in the Canton of Ticino (EMEP i, j: 70, 38; 71, 37; 71, 38) were used to calculate their ratio. Afterwards, wet deposition maps of base cations were divided by the average wet to dry deposition ratio (=14) to create dry deposition maps of base cations.

Wet deposition maps were calculated by multiplying precipitation maps with concentration maps. Slightly modified precipitations maps delivered by Meteotest were used. For the period 1988-1999 precipitation maps were supplied by Meteotest, based on a dataset of FOWG (2000), for the period 2000-2007, they were calculated by Meteotest using the same method based on monitoring data of MeteoSwiss, whereas for the period 2008-2012 the maps were prepared based on gridded data of MeteoSwiss. These maps were optimized adding precipitation data from regional meteorological networks (Steingruber, 2015).

Concentrations of sulphur, nitrogen and base cations correlated well with the 3 geographic parameters latitude, longitude and altitude (Steingruber, 2015). It was therefore possible to calculate parameters for multiple linear regression equations describing the studied concentrations as a function of the three geographic parameters and so estimate average rainwater concentrations over the area of the Canton of Ticino. To increase the significance of the multiple regression equations, also data collected at Italian sites located close to the Swiss border, next to our precipitation sampling sites, were used in the analysis (provided by CNR-ISE in Verbania Pallanza; for a description of the sites see Rogora et al., 2012). In addition, to facilitate the modeling of rainwater concentrations at very high altitudes, results from the analysis of snow sampled at the Basodino glacier (2650-3100 m) were also considered. Snow cores representing the snow fallen between October and May were sampled almost every spring since 1993. All maps have a resolution of 1 x 1 km. Maps for deposition of potential acidity were calculated by adding up maps of total nitrogen and total sulphur and subtracting base cations maps.

For a more detailed description of the mapping method, see Steingruber and Colombo (2010) and Steingruber (2015).

RESULTS

Fig. 1 shows pH values and concentrations of sulphate, nitrate and ammonium in rainwater from 1988 to 2013. The Mann-Kendall trend analyses (data not shown; Steingruber, 2015) revealed that sulphate concentrations decreased significantly at all sampling sites. Concentrations of nitrate also decreased significantly at most sites, although not as much as sulphate. Decreases in concentrations of ammonium are still difficult to observe. A statistically significant reduction can only be observed at Locarno Monti, Piotta and Stabio. Nevertheless, as a consequence of the large reduction of sulphate concentrations, rainwater acidity decreased at all sites. Rainwater pH at Locarno Monti and Lugano increased from average values around 4.3 in the 1990's to present values ranging between 5.3 and 5.6.

Because of the strong influence of precipitation volumes, temporal trends observed for concentrations are more difficult to be seen for depositions (Fig. 2). In fact, the Mann-Kendall trend analysis showed again significant decreasing trends in sulphate deposition at almost all sites, while significant decreasing trends in nitrate deposition are detected only at 4 sites (instead of 6) and of ammonium only at Locarno Monti (instead of 3 sites).

Temporal changes of total sulphur and total nitrogen loads are in line with observations of wet deposition of sulphate, nitrate and ammonium. Total deposition of sulphur decreased considerably during the monitoring period (fig. 4). Average total sulphur deposition in the Canton of Ticino decreased from 111 meq m^{-2} yr^{-1} (1988-1992) to 38 meq m^{-2} yr^{-1} (2008-2012). Deposition of nitrogen also generally decreased, but this trend is still disturbed by precipitation volumes. During particularly wet periods (e.g. 1998-2002, fig. 3), values can increase again to levels similar to those observed at the beginning of the monitoring period (fig. 5). In fact, average total deposition of nitrogen during the different time periods were 152, 125, 148, 117 and 126 meq m^{-2} yr^{-1}, respectively. Oxidized (Nox) and reduced (Nred) nitrogen contributed with about 50% each to the total.

The stronger reduction in total deposition of sulphur compared to nitrogen caused also a change in the relative contribution of sulphur and nitrogen to the total acidifying load (tab. 1). During 1988-1992, 41% of the acidifying load was determined by sulphur and 59% by nitrogen compounds, whereas at present the contribution of sulphur has almost halved (23% sulphur and 77% nitrogen).

A further result of the analysis was that, for both sulphur and nitrogen, wet deposition contributed most to total deposition. From 1988 to 2012, the deposition of acidifying compounds was determined between 67% and 79% by wet deposition (tab. 1).

As a consequence of reduced sulphur and nitrogen deposition, the average load of potential acidity decreased from 114 to 54 meq m^{-2} yr^{-1} (fig. 6).

Next to temporal trends, similarly to rainwater concentrations, deposition maps also revealed strong spatial variations in deposition. Highest deposition of total sulphur, nitrogen and potential acidity occurred at low latitudes and altitudes.

Period	Oxidized sulphur		Oxidized nitrogen		Reduced nitrogen	
	wet	dry	wet	dry	wet	dry
1988-1992	26%	15%	21%	9%	21%	8%
1993-1997	27%	11%	21%	10%	22%	9%
1998-2002	27%	5%	24%	9%	28%	7%
2003-2007	19%	6%	24%	13%	28%	10%
2008-2012	21%	2%	27%	11%	29%	10%

Table 1: Relative contribution of wet and dry nitrogen and sulphur deposition to total acidifying load.

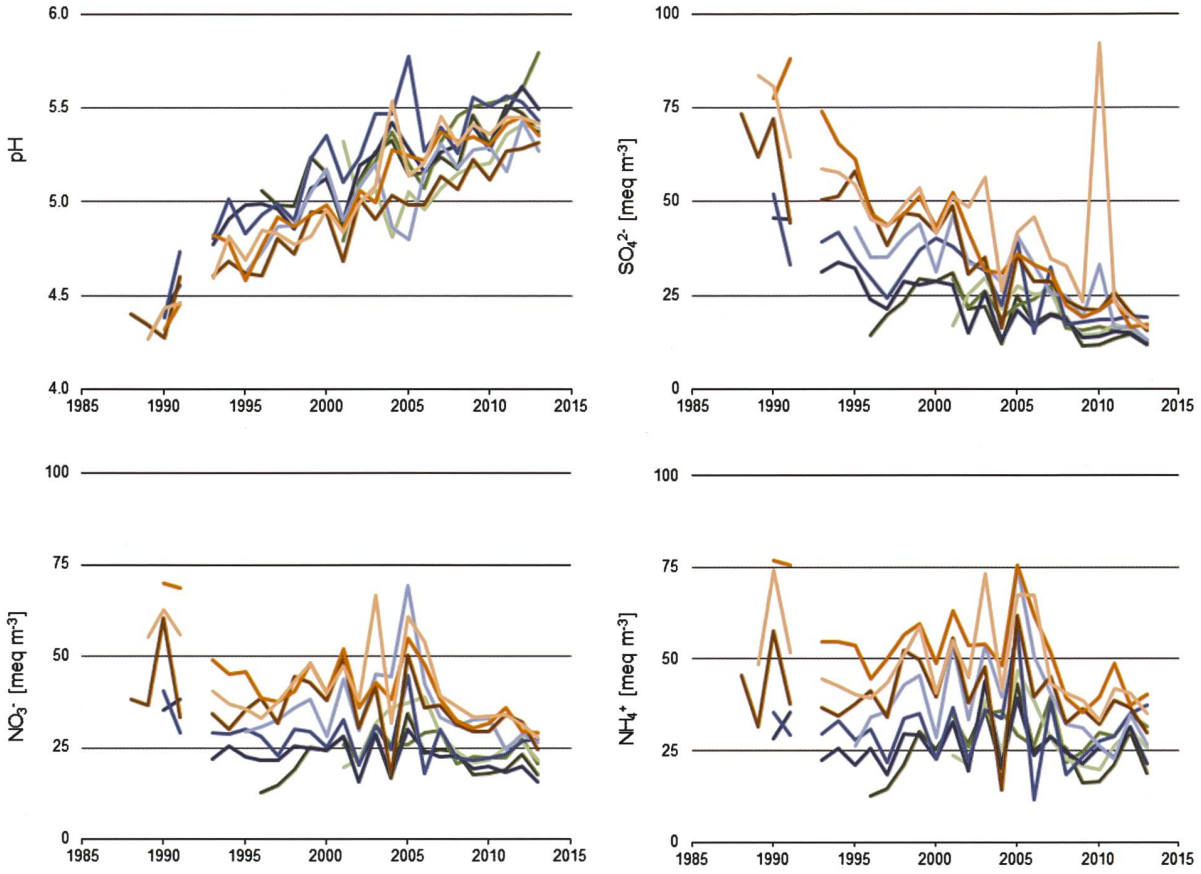

Figure 1: Long-term trends of yearly average values of pH and concentrations of sulphate, nitrate and ammonium in rainwater.

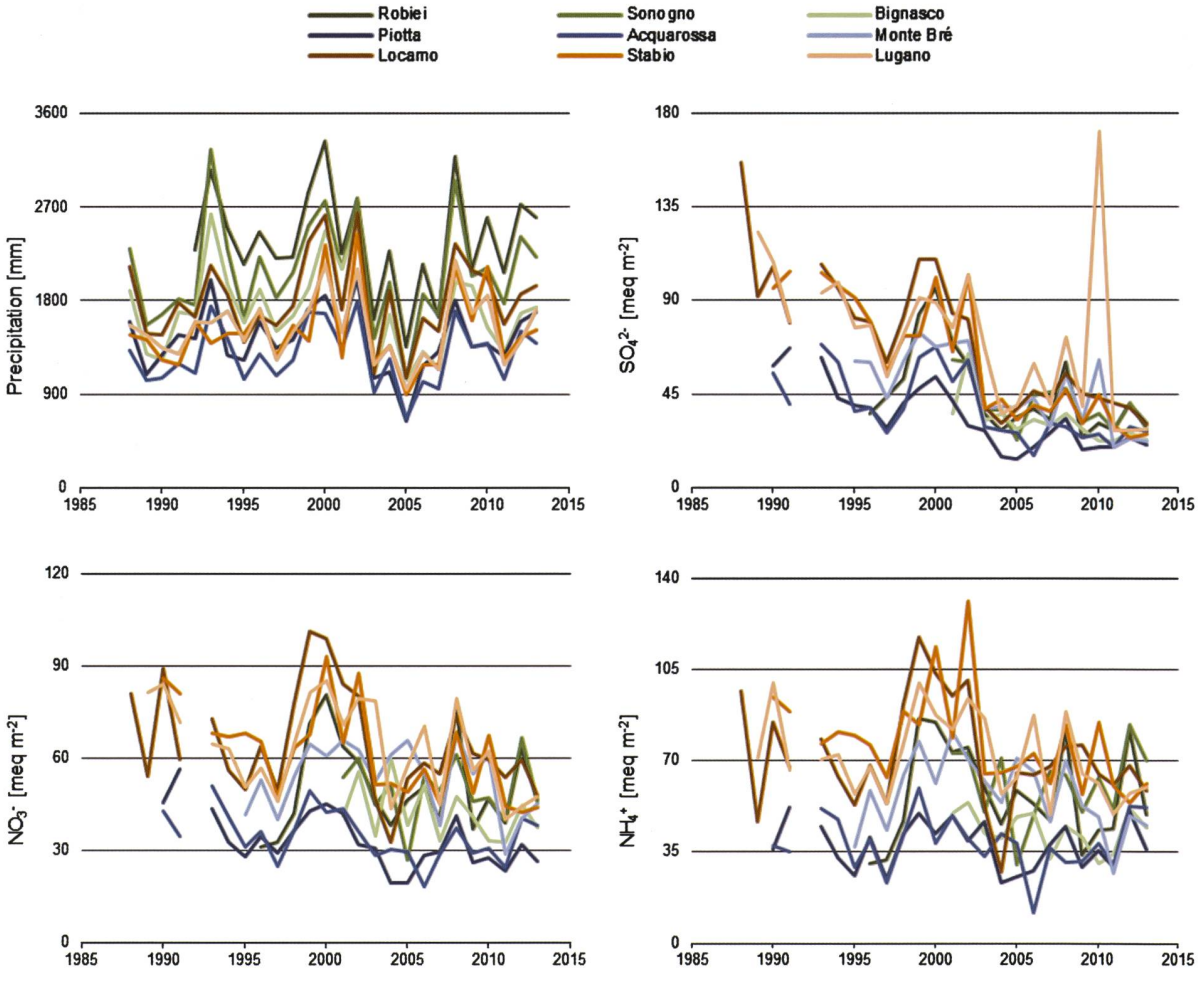

Figure 2: Long-term trends of yearly average precipitation and wet deposition of sulphate, nitrate and ammonium (legend ses Fig. 1).

DISCUSSION

The trend analysis showed a significant decrease of both rainwater concentrations and depositions for sulphate and nitrate at most sites and for ammonium at a few sites. This reflects the decrease of emission reductions. Between 1990 and 2012, at Acquarossa, Locarno Monti, Lugano, Piotta and Stabio wet deposition of sulphate decreased by 58%, 65%, 51%, 72% and 76%, of nitrate by 25%, 18%, 27%, 41% and 38% and of ammonium by 4%, 7%, 18%, 11% and 27%, respectively. However, besides these general time trends, interannual variations in rainwater concentrations and depositions also occur. The sulphate peak at Lugano in 2010 was the consequence of the volcanic eruption at Eyafjellajokull (Iceland) in April 2010. Similarly, the here not shown base cations and alkalinity peaks at Acquarossa, Locarno Monti and Piotta in 2000 and at Monte Bré, Locarno Monti, Lugano and Stabio in 2002 were caused by particularly strong alkaline rain events in October 2000 and November 2002. Precipitation also influences rainwater concentrations. In fact, because of the dilution effect of rainwater, years with lower precipitation volumes have generally slightly higher average concentrations of sulphate, nitrate and ammonium than years with higher rainwater volumes. However, much more than concentrations, precipitation volumes influence wet deposition. This is very well visible looking at the high deposition values during the rather rainy period 1999-2002, followed by the low values during the dry period 2003-2007 (fig. 2).

Similarly to rainwater concentrations and depositions, total deposition also decreased. On average, over the whole area of the Canton of Ticino, between 1990 and 2012, total deposition of sulphur decreased by 66%, oxidized nitrogen by 22% and reduced nitrogen by 13%. Interestingly, mean deposition values over the Canton of Ticino of sulphur and oxidized nitrogen are still higher than the Swiss averages. In Switzerland, between 2008 and 2012, average total deposition of sulphur, oxidized nitrogen and reduced nitrogen were 15, 30 and 68 meq m^{-2} yr^{-1}, respectively, according to European-wide modeling by EMEP (Gauss et al., 2014), whereas in the Canton of Ticino the respective values were 38, 61 and 65 meq m^{-2} yr^{-1}. In the Canton of Ticino itself total depositions also vary spatially. Similarly to rainwater concentrations they depend strongly on latitude and altitude (fig. 4-6). The correlation with latitude and altitude reflects the influence of long-range transboundary air pollution moving along a south to north gradient from the Po plain toward the Alps and the distance from pollution sources.

Differences between the Canton of Ticino and Switzerland can be observed also when considering the origin of depositions. In 2012, in Switzerland 81% of sulphur, 85% of oxidized nitrogen and 34% of reduced nitrogen deposition came from emission sources abroad (Gauss et al., 2014). For the Canton of Ticino a detailed analysis has not been made, but EMEP modeling results show that for sulphur and nitrogen the transboundary contribution to total deposition is significantly higher in Southern Switzerland than in other parts of the country (Gauss et al 2014).

Interesting is also the different contribution of sulphur and nitrogen compounds to the total acidifying load. During the most recent monitoring period (2008-2012), in the Canton of Ticino total sulphur, oxidized nitrogen and reduced nitrogen contributed on average by 23%, 38% and 39%, respectively, to the total load of acidity, while Switzerland's averages were 15%, 26% and 59% (Gauss et al., 2014). Different is also the contribution of wet and dry deposition to total nitrogen deposition. Between 1988 and 2012, in the Canton of Ticino the contribution of wet to total nitrogen deposition was on average 71%. At the Northern side of the Alps, because of lower precipitation volumes, this percentage is smaller. Thimonier et al. (2005) determined a contribution of 40-60% between 1995 and 2001 in the Jura and the Central Plateau and EKL (2005) reported an average Swiss value for the year 2000 of 51%.

In the framework of the Convention on Long-range Transboundary Air Pollution (UNECE), empirical critical loads of nitrogen (CLN) have been defined for natural and semi-natural ecosystems (Bobbink & Hettelingh, 2011). The most sensitive ecosystems in Switzerland are raised bogs with CLN of 7 kg N ha^{-1} yr^{-1} (= 50 meq m^{-2} yr^{-1}) and (sub)-alpine grasslands with CLN of 8 kg N $ha^{-1}yr^{-1}$ (= 57 meq m^{-2} yr^{-1}) (Achermann et al., 2011). For forest ecosystems in Southern Switzerland CLN of 10 kg N $ha^{-1}yr^{-1}$ (71 meq m^{-2} yr^{-1}) were modelled with the steady sate mass balance method (EKL, 2005; Rihm, 1996; Achermann et al., 2011). The exceedances of CLN can be obtained by subtracting CLN from total nitrogen deposition at sites with sensitive vegetation. FOEN (in prep.) calculated maps of CLN exceedances for Switzerland with a resolution of 1 km x1 km grid. In the Canton of Ticino, the nitrogen load exceeds the CLN at most sensitive sites. In 1990 95% of the sensitive sites had positive exceedance values, ranging between 1 and 39 kg N $ha^{-1}yr^{-1}$ (mean 14 kg N $ha^{-1}yr^{-1}$), while in 2010 the same percentage decreased to 91%, with values ranging between 1 and 29 kg N $ha^{-1}yr^{-1}$ (mean 9 kg N $ha^{-1}yr^{-1}$). These results indicate that the exceedance of CLN is still a problem in Southern Switzerland. It has been estimated that non-exceedance of CLN can be reached by reducing NO_x and NH_3 emissions of 2005 by 50% and 40%, respectively (BAFU, 2009); obviously this result can be achieved only if neighbor countries reduce their emissions by the same percentage. Between 2005 and 2012 emissions of nitrogen compounds in Switzerland have been reduced by 17% (36% NO_x, 6% NH_3; Heldstab et al., 2014). For the Canton of Ticino only the reduction in NO_x emissions is known: 20% (UACER, 2007).

Compared to nitrogen deposition, the situation looks better for deposition of acidity. Critical loads of acidity (CLA) for Swiss forests soils have been calculated with the first-order acidity balance model (Achermann et al., in prep). During 1988-1992, 81% of the national forest inventory sites had depositions of nitrogen and sulphur that exceeded critical loads. The same percentage decreased to 67% in 1993-1997, 72% in 1998-2002, 40%

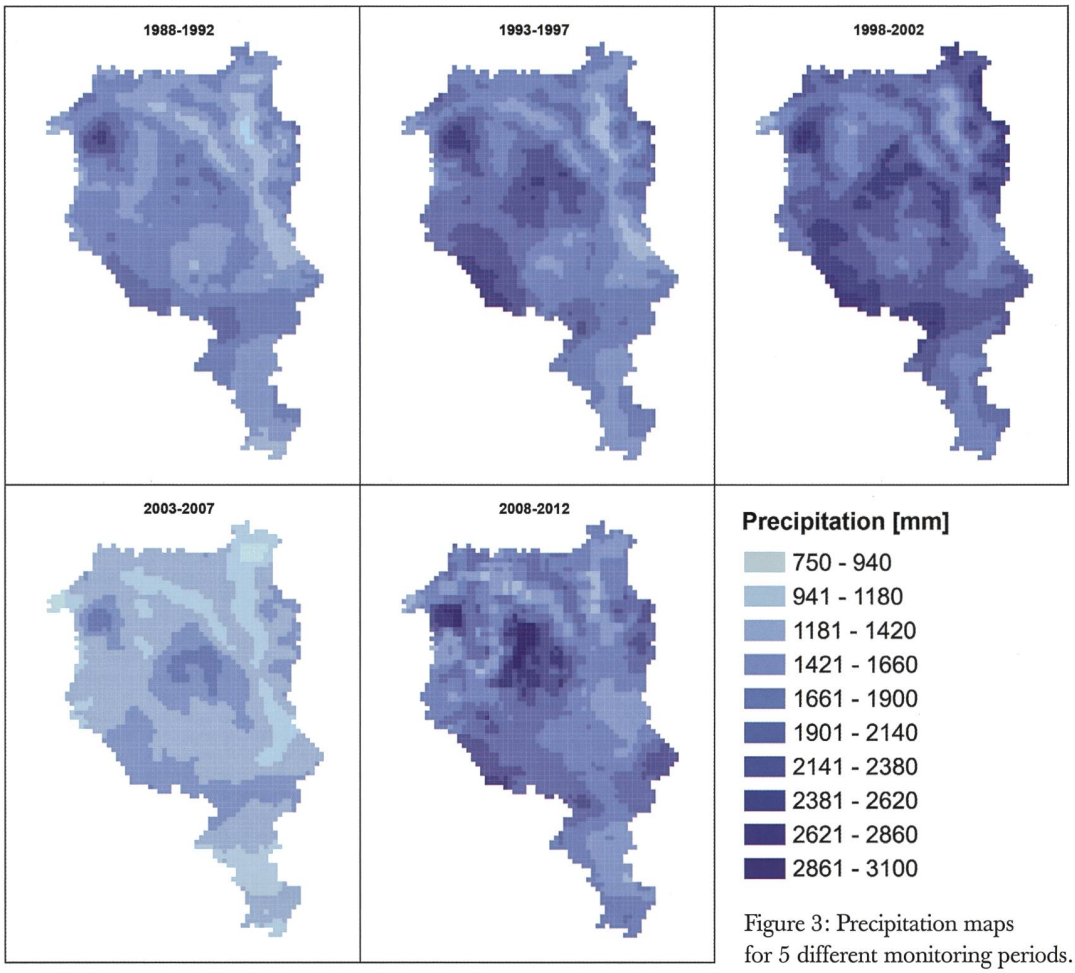

Figure 3: Precipitation maps for 5 different monitoring periods.

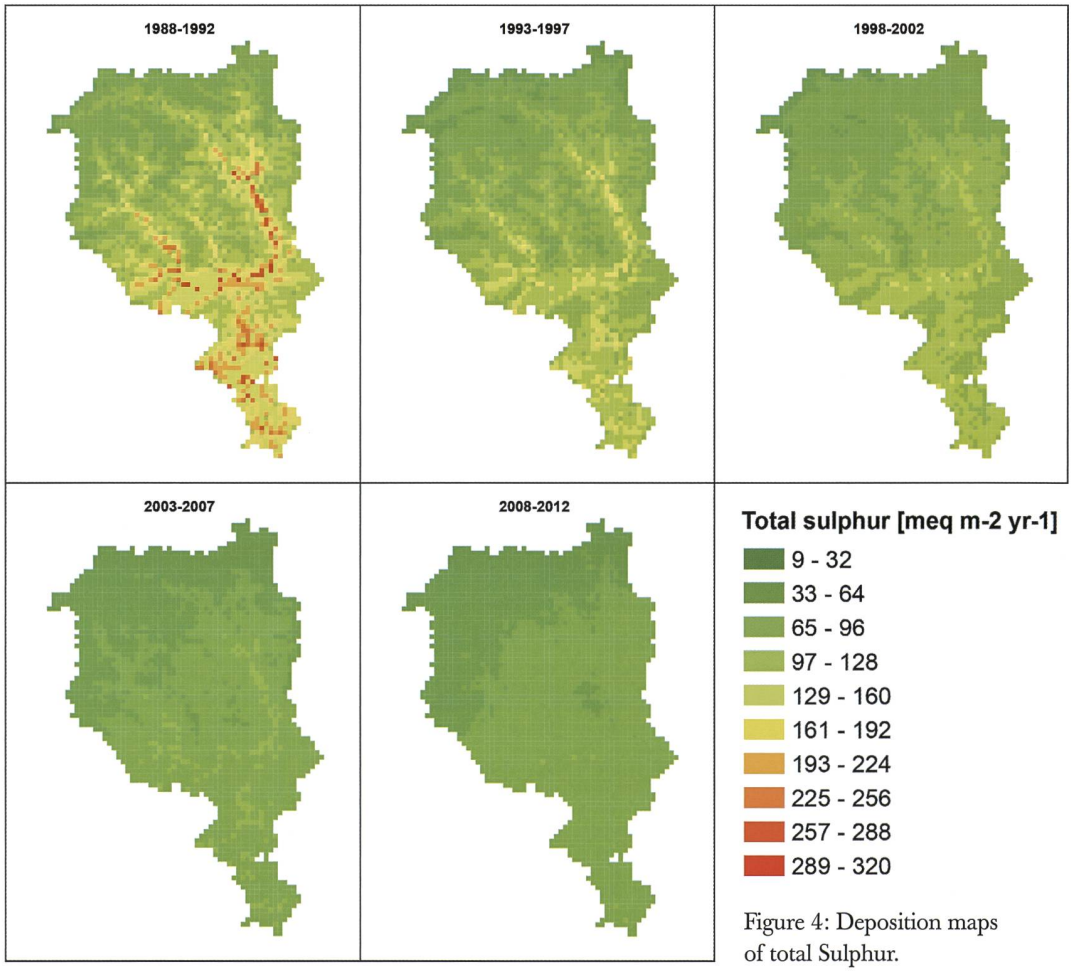

Figure 4: Deposition maps of total Sulphur.

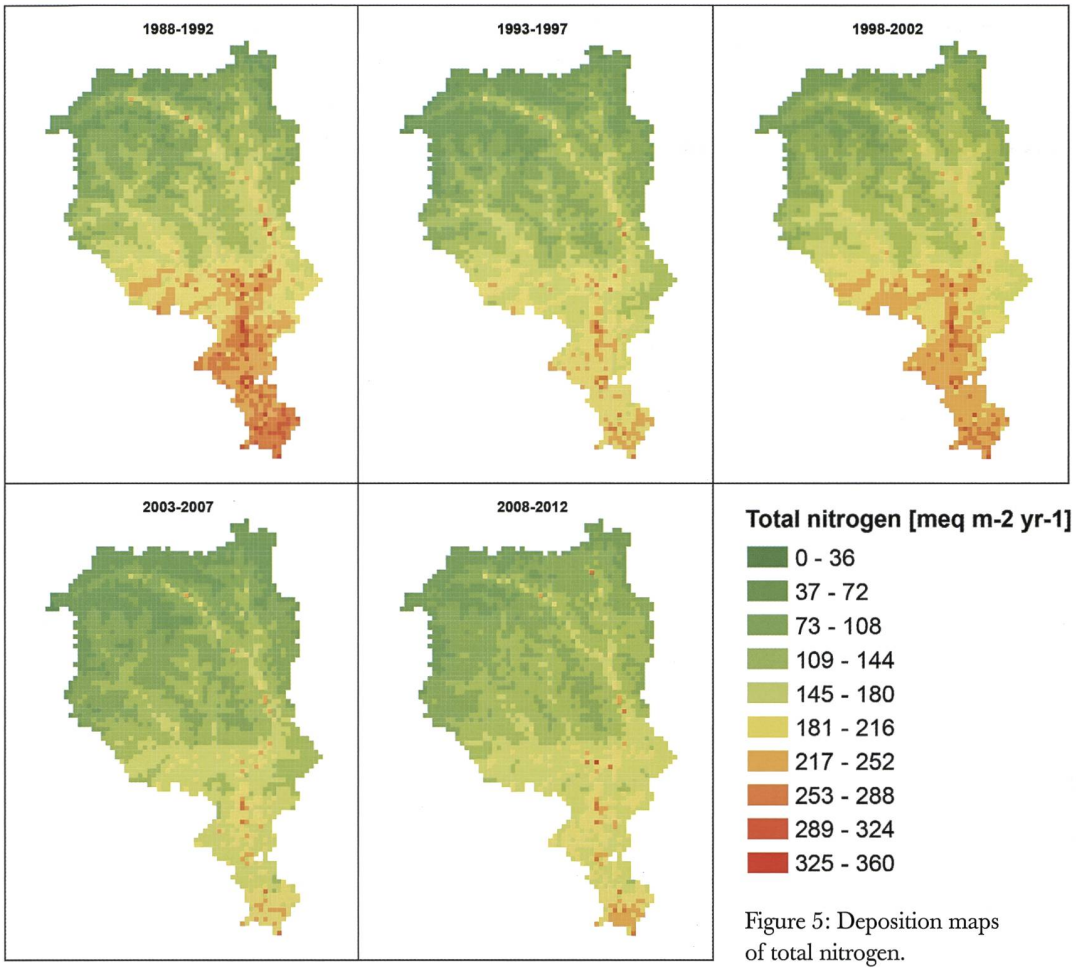

Figure 5: Deposition maps of total nitrogen.

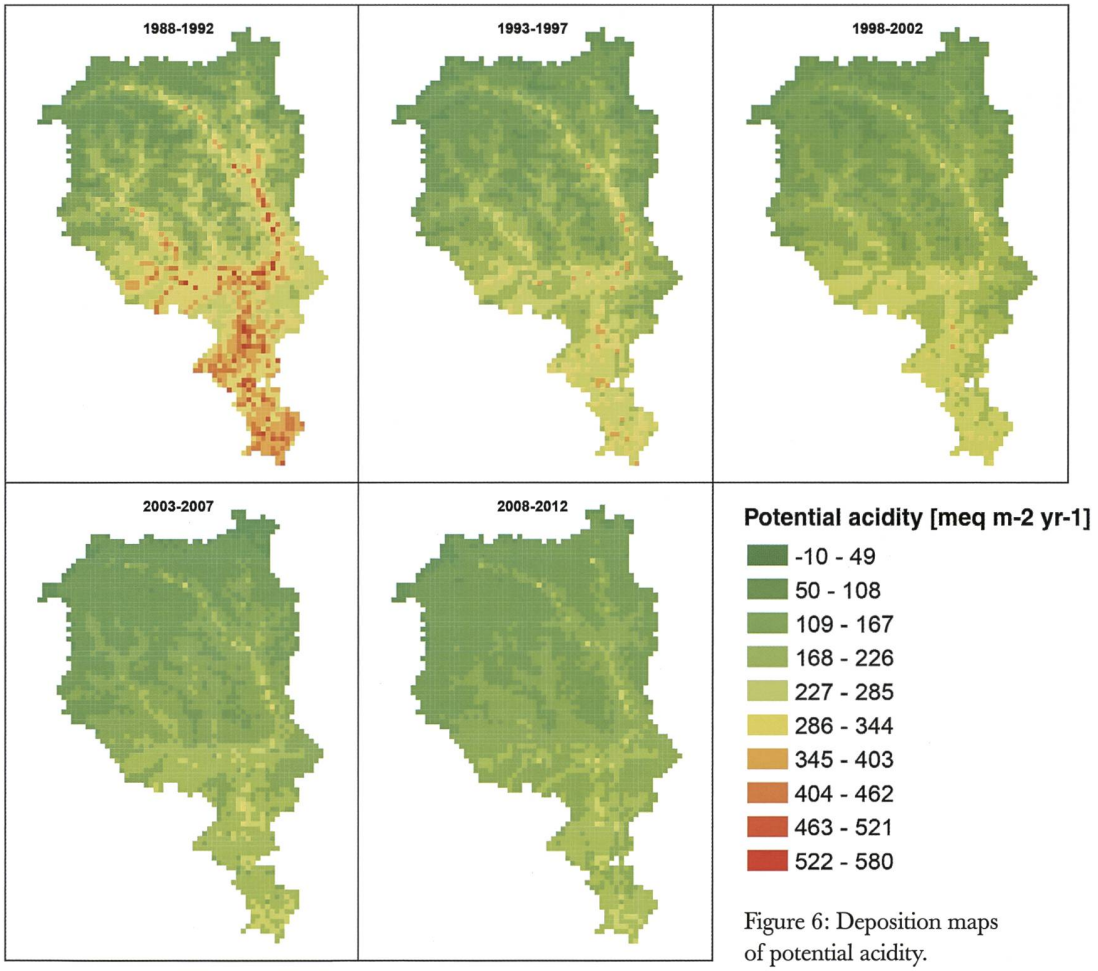

Figure 6: Deposition maps of potential acidity.

in 2003-2007 and 43% in 2008-2012. Similar results were obtained for alpine lakes. At the beginning of the monitoring period, acid neutralizing capacity (ANC) measured in lake surface water in autumn was below the critical limit of 20 meq m^{-3} (Posch et al., 2007) in 13 lakes out of 20, while at present only 6 lakes have ANC values below 20 meq m^{-3} and 3 of them have ANC values very close to the acceptable limit (Steingruber & Colombo 2006, 2014).

To conclude, the Canton of Ticino, compared to the rest of Switzerland is particularly exposed to deposition of atmospheric pollutants because of its proximity to the Po Plain in Italy, an area with high anthropogenic emissions, and because of its rainy climate. In addition, since base poor rocks, mainly gneiss, dominate most of the lithology in Southern Switzerland, many high altitude surface water bodies with small catchments and forest soils are sensitive to acidification. While deposition of acidifying pollutants is nowadays below the critical load at most sensitive sites, deposition of nitrogen is still too high. Further emission reductions of both NO_x and NH_3 in Switzerland and in its neighboring countries are therefore necessary.

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