Acidifying Deposition in Southern Switzerland

Monitoring, maps and trends 1983-2017

Ufficio dell'aria, del clima e delle energie rinnovabili
Sandra Steingruber
Telefono: 091 814 29 30, fax: 091 814 29 79 e e-mail: sandra.steingruber@ti.ch
Bellinzona, 15.11.2018

Content

Abstract

Sulphur and nitrogen oxides from combustion processes and ammonia from agriculture can be transported over long distances, transformed and then loaded on natural ecosystems causing acidification and eutrophication of sensitive ecosystems. Because of its proximity to the emission rich Po Plain and its generally abundant precipitations, Southern Switzerland is particularly exposed to deposition of anthropogenic pollutants.

Total sulphur, nitrogen and acid deposition maps for the Canton of Ticino for the seven time periods 1983-1987, 1988-1992, 1993-1997, 1998-2002, 2003-2007, 2008-2012, 2013-2017 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 during the last 30 years average total deposition of sulphur and nitrogen decreased from 114 to 25 meq m^{-2} yr^{-1} and from 158 to 117 meq m^{-2} yr^{-1}, respectively. As a consequence of reduced sulphur and nitrogen deposition, the average present load of acidity decreased from from 202 to 104 meq m^{-2} yr^{-1}.

The analysis also showed that most deposition of acidifying compounds occurs through wet deposition (71-79%). As a consequence of the strong decrease in sulphur deposition the relative importance of sulphur compounds in determining total deposition of acidifying compounds has decreased from 42% to 18%, while that of nitrogen compounds has increased from 58% to 82%, with oxidized and reduced nitrogen contributing about 50% each to the total.

For illustration of a terrestrial ecosystem, average exceedances of critical loads of acidity and nutrient nitrogen have been calculated for forest sites. Between 1985 and 2015 these exceedances decreased from 134 to 30 meq m^{-2} yr^{-1} and from from 113 to 66 meq m^{-2} yr^{-1}, respectively. While the percentage of forest sites with exceeded critical load of acidity decreased significantly from 81% to 26%, the percentage with exceeded critical load of nutrient nitrogen remained unchanged (99%).

Introduction

 "Acid rain" is a broad term used to describe the deposition pathway of acidifying compounds from the atmosphere to the surface of the earth. Acidifying deposition has two components: wet and dry. Wet deposition refers to acidifying rain, fog, and snow, while dry deposition refers to acidifying gases and particles. The primary causes of acidifying deposition are the emissions of sulphur dioxide (SO_2) and nitrogen oxides (NO_x) from combustion of fossil fuels as well as ammonia (NH_3) emissions from agriculture. In the atmosphere SO_2 and NO_x can be oxidized to sulphuric and respectively nitric acid causing acid precipitation. Although ammonia itself reacts as a base in the atmosphere (resulting in the formation of ammonium, NH_4^+), during the assimilation by plants the temporary bound proton is released again to the environment. In addition, in soils and waters ammonium can be oxidized by microorganisms to nitrate (nitrification), releasing two protons. In this way, ammonia emissions can contribute to the acidification of soils and waters.

Acidifying deposition affects the environment in several ways. Acidification of surface waters gradually leads to severe changes in biological communities. Effects range from reductions in diversity without changes in total biomass to elimination of all organisms (Dillon et al. 1984). Damages to forests include weakening of the root system, nutrient imbalances and defoliation. Building materials and works of art can also be damaged by corrosion due to acid deposition. Also health problems, especially respiratory and cardiovascular diseases, have been found to be associated with increased concentrations of particulate matter (i.e. aerosols) and ozone, both formed by precursors such as sulphur oxides, nitrogen oxides, volatile organic compounds and ammonia.

Acidifying deposition first began with the industrial revolution, when large amounts of fossil fuels were burnt to produce steam power needed to drive machinery. The term "acid rain" was coined in the 19th century by the scientist Robert Smith, working at the time in Manchester (Smith 1852). In those times acid rain was confined to industrial towns and cities. However, the situation gradually worsened and widespread environmental damage on a global scale was observed by scientists in the second half of the 20th century.

In the sixties the link between sulphur emissions in continental Europe and acidification of Scandinavian lakes had been demonstrated (Odén 1968). Between 1972 and 1977 several studies confirmed the hypothesis that air pollutants can travel several thousands of kilometers before deposition and damage occur, evidencing that cooperation on an international level was necessary to solve problems such as acidification. As a consequence in 1979 34 Governments, including Switzerland, and the European Community (EC) signed the Convention on Long-range Transboundary Air Pollution (CLRTAP). The Convention entered into force in 1983. Today it has 51 Parties and has been extended by eight specific protocols. Four of these protocols control acidifying pollutants.

The Helsinki Protocol of 1985 aimed at reducing sulphur emissions by at least 30%. The goal of the 1988''s Sofia Protocol was the freezing of the emissions of NO_x. The 1994's Oslo Protocol required further reduction of sulphur emissions and the Gothenburg Protocol of 1999 set national emission ceilings for sulphur, NO_x, VOC's and ammonia for 2010. As a consequence, a substantial reduction in the emissions of sulphur and nitrogen oxides

(EMEP 2016) has been achieved over the last 20-25 years leading to an improved quality of atmospheric deposition.

Figure A: Annual sulphur dioxide, nitrogen oxides and ammonia emissions in Switzerland from 1900 to 2030. Dashed lines after 2016 are projections.
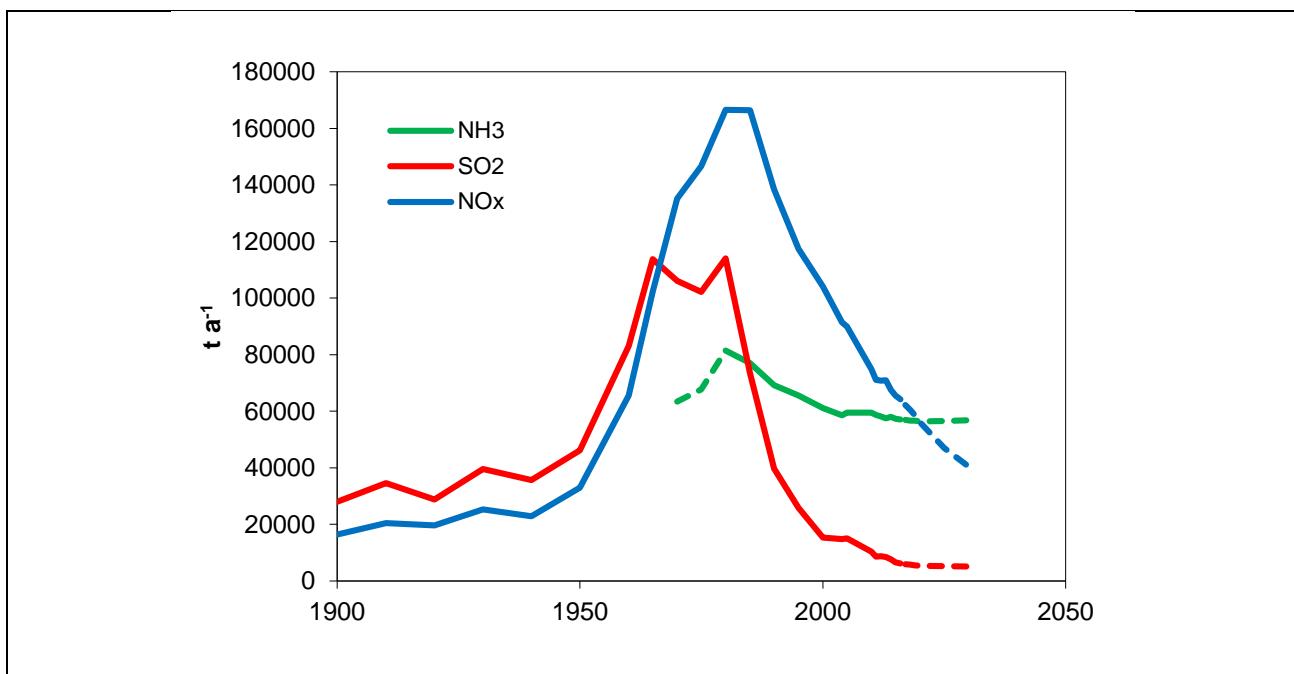

Fig. A shows the emissions of sulphur and nitrogen oxides and ammonia in Switzerland from 1900 to 2030 (Heldstab et al. 2018). The sulphur and nitrogen oxides emissions started to increase steeply after the second world war. Sulphur oxides reached their maximum between 1965 and 1980, while nitrogen oxides peaked around 1985. Afterwards, both sulphur and nitrogen oxides decreased continuously until present (2016). Ammonia decreased only little. The reduction of sulphur dioxide emissions has mainly been caused by a reduction of the sulphur content in liquid fuels and the partial substitution of sulphur rich coal with other fossil fuels. The decrease of the nitrogen oxides emissions after 1985 has been mainly determined by the equipment of cars with catalytic converters and stationary combustion sources with DeNO$_x$-systems. However, 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 and particularly from the heavily polluted urban area of Milan and Turin. 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. Furthermore, high altitude soils and freshwaters of southern Switzerland are particularly sensitive to acidification because of the dominance of base-poor rocks with low buffering capacity. In the recently published ICP waters report 135/2018 (Austnes et al. 2018) Steingruber showed that at present (2015-2017) in Switzerland still 25% of the analyzed potentially acid sensitive lakes (52) have autumn ANC values below 20 meq m^{-3} and 10% of the same lakes have pH values below 6.0. Compared to the past, the present acidification status has improved. During the large scale survey in 1995, 40% and 29% of the 45 analyzed lakes had autumn ANC and pH values below 20 meq m^{-3} and 6.0,

respectively. Decreasing depositions of S have been the main reason for the observed chemical recovery (Rogora et al. 2013). However, at present N accounts for about 80% of the acidifying deposition (Rogora et al. 2016), which mean that for further recovery of lake chemistry emissions of N have to be significantly reduced. The same report concludes that acidification is still observed in many other countries in Europe and North America, as well and that even by reaching the emission targets of acidifying compounds set for 2030, critical loads for surface waters will remain exceeded. As a consequence, it is important to continue to monitor acidifying deposition, especially at the more sensitive sites. As regards southern Switzerland, acidifying deposition has already been assessed by Barbieri and Pozzi (2001), Steingruber and Colombo (2010) and Steingruber (2015) for the following time periods: 1988-1992, 1993-1997, 1998-2002, 2003-2007, 2008-2012. This report includes an update of all already published 5-years deposition maps, maps of the most recent period (2013-2017) and for the first time deposition maps estimated for the most polluted period (1983-1987). In particular, the aims of this report are:

➢ to describe rainwater quality at different sampling stations in southern Switzerland from 1988 to 2017;
➢ to calculate temporal trends for the main chemical parameters present in rainwater involved in the process of acidification;
➢ to map wet deposition of the main chemical parameters for southern Switzerland for five-years periods from 1983 to 2017 with the aid of multiple regression analysis between concentrations of parameters relevant for acidification and geographic parameters;
➢ to map total deposition by adding up wet and dry deposition, the latter being modeled by Meteotest.

The here calculated rainwater concentration models for southern Switzerland were then integrated in the Swiss deposition maps developed under request of the Swiss Federal Office for the Environment (https://www.bafu.admin.ch/bafu/de/home/themen/luft/zustand/daten/luftbelastung--historische-daten/karten-jahreswerte/karte-stickstoff-deposition.html).

1. Precipitations in Southern Switzerland

1.1 Introduction

Precipitation volumes influence very much rainwater quality and the amount of wet deposition of air pollutants. For this reason precipitation maps from 1983 to 2017 have been calculated in 5-years time periods.

1.2 Sampling sites

Yearly precipitation from totally 96 pluviometric stations were used to estimate the amount of precipitation over southern Switzerland. Swiss data originated from different precipitation monitoring networks: the Federal Office of Meteorology and Climatology (MeteoSwiss), the Canton of Ticino with data from Ufficio dei corsi d'acqua (UCA) and from Ufficio del monitoraggio ambientale (OASI). At one station the amount of precipitation was measured by the Federal Institute for Forest, Snow and Landscape Research (WSL). Italian data were provided by the Institute of Ecosystem Study (ISE-CNR), the Regional Agencies for the Protection of the Environment of Piedmont and Lombardy (ARPA Piemonte and ARPA Lombardia), the national agency for electric energy (ENEL) and the hydroelectric power agencies (Idroelettriche Riunite S.p.A.). The geographic distribution of the precipitation sampling sites is shown in Fig. 1.1. Longitudes, latitudes, altitudes, data source are reported in Tab. A1 of the Appendix.

Figure 1.1: Precipitation sampling sites. Swiss sites: red (MeteoSwiss), green (UCA), orange (OASI), black (WSL); Italian sites: blue.
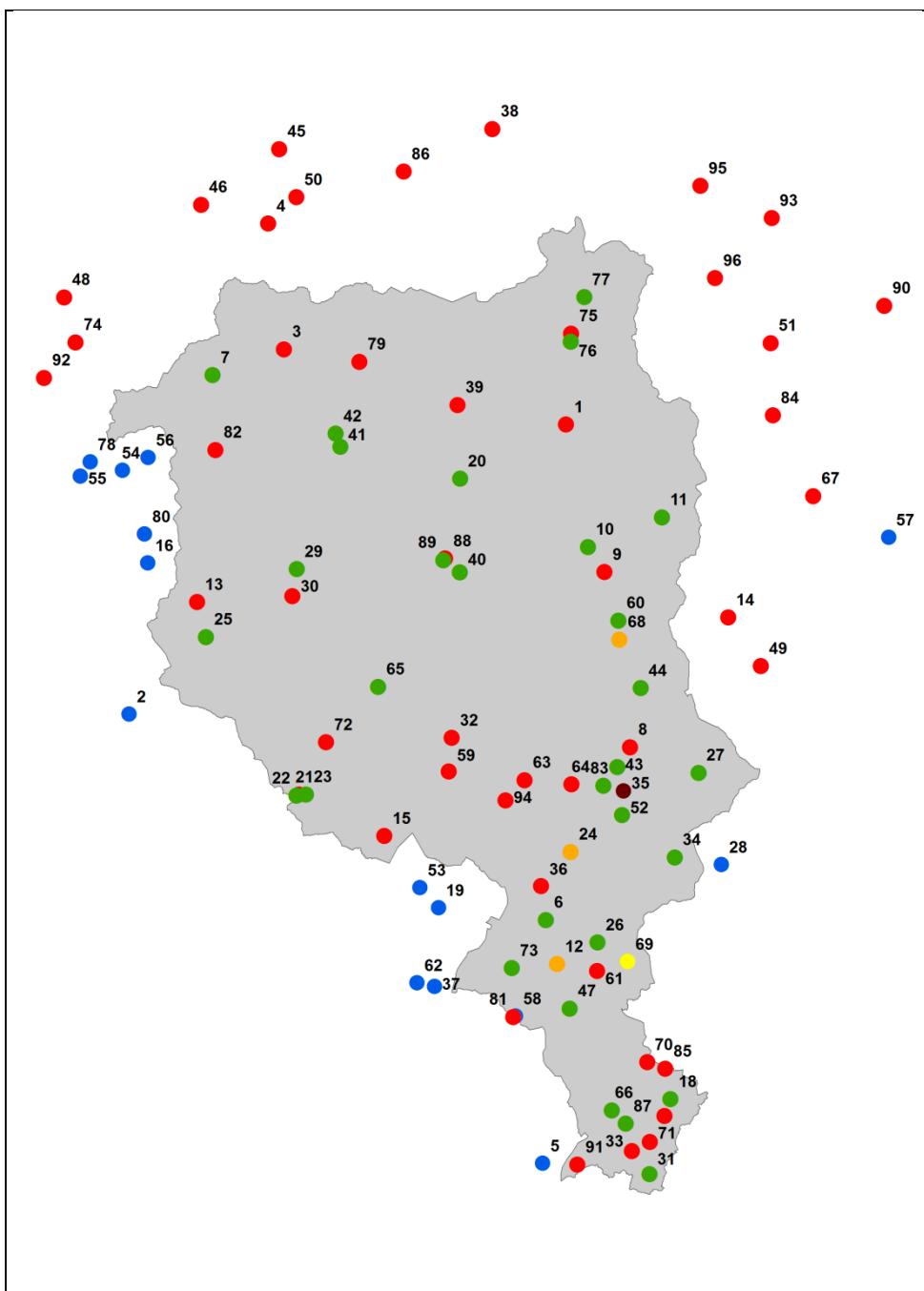
1.3   Mapping method

Existing national precipitation maps were refined for the study area with the following procedure: for each of the 96 sampling stations listed in Tab. A1 of the Appendix the average precipitation volumes over all of the available five years periods of 1983-1987, 1988-1992, 1993-1997, 1998-2002, 2003-2007, 2008-2012, 2013-2017 were calculated and divided by values extracted from national precipitation maps (resolution: 1km x 1km). For the period of 1983-1999 the precipitation maps were supplied by Meteotest based on a dataset of FOWG (2000), whereas for the period 2000-2007, they were calculated by the company Meteotest using the same method based on monitoring data of MeteoSwiss. For the period 2008-2017 the maps were prepared based on gridded data of MeteoSwiss.

The resulting factors were interpolated by the inverse distance weighting method in ArcGIS® (registered trademark of Esri Inc., Redlands, USA) using the following parameters: distance exponent = 2, number of points = 3, maximal search distance = 11 km, resolution = 1 km x 1km. These maps were then multiplied back by the precipitation maps.

1.4   Precipitation maps

The calculated precipitation maps are shown in Fig. 1.2. Mean annual precipitation was 1903 mm in 1983-1987, 1667 mm in 1988-1992, 1873 mm in 1993-1997, 2038 mm in 1998-2002, 1313 mm in 2003-2007, 1880 mm in 2008-2012 and 1815 mm in 2013-2017. Interestingly, 1998-2002 was one of the wettest and 2003-2007 one of the driest 5-year period ever measured. The wettest region is situated in the western part of the study area. This region includes the Centovalli's, the Onsernone's and the lower Maggia's valley. The reasons for this distribution are air masses rich in humidity moving predominantly from southwest toward the southern Alps and the particular orography of the area causing a steep raise of the air masses to higher altitudes. Other rain rich regions are located in the northwestern part (higher Maggia valley), in the north-central part (higher Verzasca valley) and in the centre of the Canton of Ticino (mount Tamaro-Gradiccioli). Precipitation is lowest in the eastern part of the Canton due to less frequent exposure to humid currents. For a more detailed description of the climate in the studied area one may refer to Spinedi and Isotta (2004) and MeteoSvizzera (2012).

Figure 1.2: Precipitation maps: 1983-1987, 1988-1992, 1993-1997, 1998-2002, 2003-2007, 2008-2012, 2013-2017
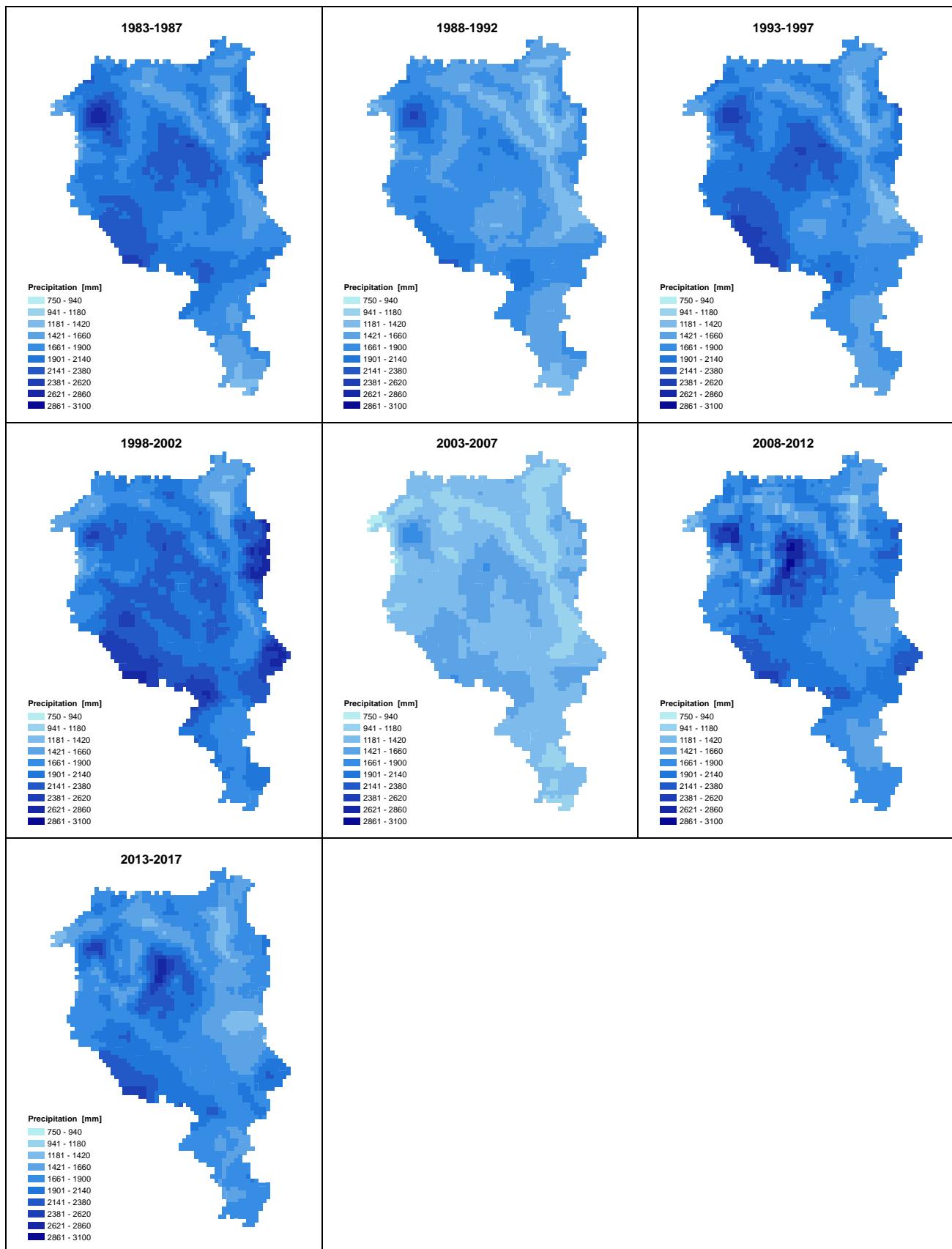
2. Rainwater quality

2.1 Sampling sites

Sampling of wet deposition was carried out at weekly intervals. Between 1982 and 1985 rainwater was collected at Locarno Monti and Lugano with bulk samplers. Since 1988, wet-only samplers have been used. Sampling of wet deposition started at Acquarossa, Piotta and Stabio in 1990, at Monte Bré in 1995, at Robiei in 1996, at Bignasco and Sonogno in 2001. Sampling sites were chosen along a south-north axis and at various altitudes (200-1900 m a.s.l.). In order to better describe the dependence on geography, results from the closed-by Italian sampling sites have been also considered in the statistical analysis (data have been provided by the Institute of Ecosystem Study in Pallanza, Italy). 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. The geographic distribution of the sampling sites and their geographic coordinates are shown in Fig. 2.1 and Tab. 2.1, respectively.

Figure 2.1: Study area with wet deposition sampling points. Swiss: red, Italian: blue.
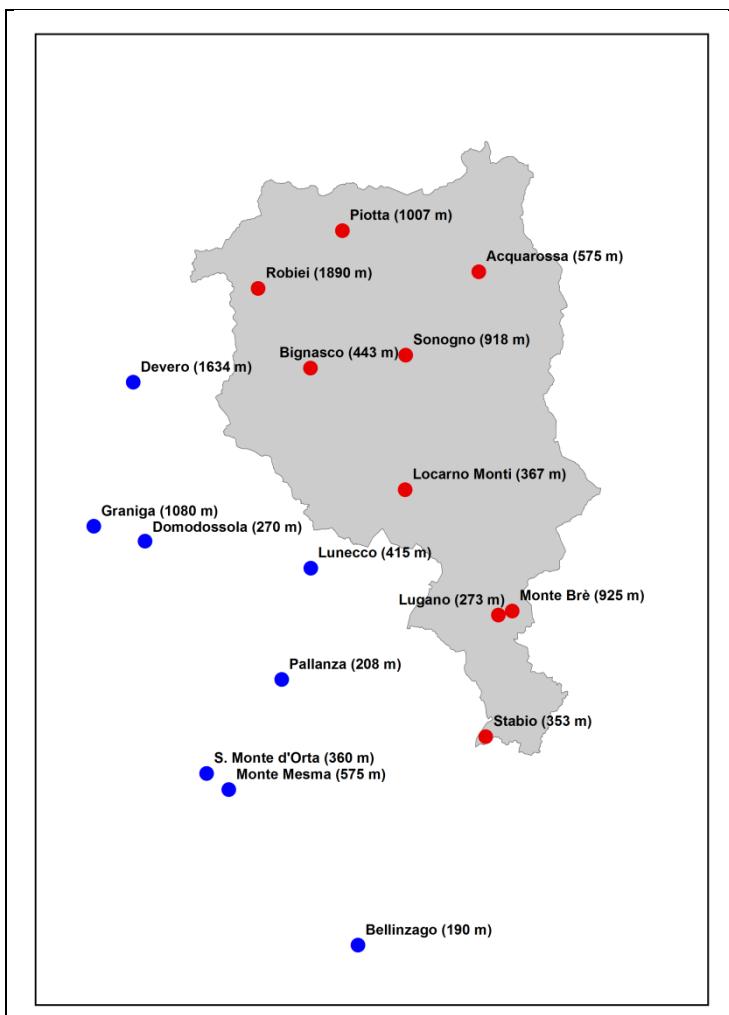
Table 2.1: Swiss (CH) and Italian (I) wet deposition sampling sites and their geographic (WGS84) and Swiss (CH1903 LV03) coordinates, altitudes and sampling years

Sampling site	WGS84		CH1903 LV03 (m)		Altitude (m a.s.l.)	Sampling years
	North	East	North	East		
Acquarossa (CH)	46°27'41''	8°56'12''	146440	714998	575	1990-1991, 1993-2017
Bignasco (CH)	46°21'11''	8°36'41''	132257	690205	443	2001-2017
Locarno Monti (CH)	46°10'27''	8°47'17''	114350	704160	367	1982-1985, 1988-1991, 1993-2017
Lugano (CH)	46°00'24''	8°57'18''	95870	717880	273	1982-1985, 1989-1991, 1993-2017
Monte Brè (CH)	46°00'32''	8°59'17''	96470	719900	925	1995-2017
Piotta (CH)	46°31'07''	8°40'35''	152500	694930	1007	1990-1991, 1993-2017
Robiei (CH)	46°26'43''	8°30'51''	143984	682540	1890	1996-2017
Sonogno (CH)	46°21'05''	8°47'14''	134150	704250	918	2001-2017
Stabio (CH)	45°51'36''	8°55'52''	77970	716040	353	1990-1991, 1993-2017
Bellinzago (I)	45°34'27''	8°41'31''	47245	697217	190	1989-2015
Devero (I)	46°19'19''	8°16'29''	130156	664132	1634	1996-2017
Domodossola (I)	46°06'42''	8°17'41''	106767	665875	270	1986-2017
Graniga (I)	46°07'52''	8°11'59''	108936	658342	1080	1994-2012
Lunecco (I)	46°04'28''	8°36'39''	102774	690264	415	1989-2015
Monte Mesma (I)	45°46'43''	8°26'38''	70166	678202	575	2003-2016
Pallanza (I)	45°55'42''	8°34'48''	86386	686003	208	1985-2017, 1983-2017 (bulk)
S. Monte d'Orta (I)	45°48'13''	8°24'41''	72547	674929	360	1990-1999
Basodino glacier (CH)	46°25'04''	8°28'34''	141000-141500	679500-680000	2650-3100	1993-2003, 2006-2018

2.2 Analytics

Rain samples were analyzed for pH, alkalinity, conductivity and the main cations and anions. Parameters, analytical methods and quantification limits are shown in Tab. 2.2.

Table 2.2: Measured parameters, analytical methods, accuracy and quantification limits

Parameter	Filtration	Conservation	Methods	Accuracy
pH	No	No	potentiometry	0.02
conductivity	No	No	Kolrausch bridge (20°C)	0.5 µS cm^{-1}
alkalinity	No	No	potentiometric Gran titration	0.001 meq l^{-1}
				Quantification limit
Ca^{2+}	CA filter	PP bottle, 4°C	ion chromatography	0.010 mg l^{-1}
Mg^{2+}	CA filter	PP bottle, 4°C	ion chromatography	0.005 mg l^{-1}
Na^+	CA filter	PP bottle, 4°C	ion chromatography	0.005 mg l^{-1}
K^+	CA filter	PP bottle, 4°C	ion chromatography	0.010 mg l^{-1}
NH_4^+	CA filter	PP bottle, 4°C	spectrophotometry	3 mg N l^{-1}
SO_4^{2-}	CA filter	PP bottle, 4°C	ion chromatography	0.005 mg l^{-1}
NO_3^-	CA filter	PP bottle, 4°C	ion chromatography	0.010 mg N l^{-1}
Cl^-	CA filter	PP bottle, 4°C	ion chromatography	0.010 mg l^{-1}

The quality of the data was assured by regular participation to national and international intercalibration tests. In addition, data were accepted only if the calculation of the ionic balance and the comparison between the measured and the calculated conductivity corresponded to the quality requests included in the programme manual of ICP Waters (ICP waters Programme Centre 2010).

2.3 Concentrations of chemical parameters in rainwater

Fig. 2.2 shows the yearly average concentrations of the main chemical parameters measured in precipitation sampled at the 9 Swiss sampling sites Acquarossa, Bignasco, Monte Brè, Locarno Monti, Lugano, Piotta, Robiei and Sonogno between 1988 and 2017.

The corresponding data are tabulated in Tab. A2 of the Appendix. Yearly mean concentrations were calculated by weighting weekly concentrations with the sampled precipitation volume:

$$C(X)_a = \frac{\sum_w P_w \cdot C(X)_w}{P_a} \quad \text{where}$$

P_w = weekly precipitation volume (measured with the wet-only sampler)

$C(X)_w$ = weekly concentration of compound X

P_a = annual precipitation volume calculated as sum of P_w

In addition to the temporal trends that are analyzed and discussed in chapter 2.4, it can be observed that concentrations can vary very much from one year to the other. Because of dilution, during wet years concentrations of sulphate, nitrate and ammonium tend to be lower and during dry years higher than average. It also can happen that single particularly intense rain events with alkaline characteristics can heavily influence yearly mean base cation concentrations and acidity. Exceptionally high base cations and low acidity peaks can be observed at sampling stations Acquarossa, Locarno Monti and Piotta in 2000 (alkaline event in October) and at Monte Bré, Locarno Monti, Lugano and Stabio in 2002 (alkaline event in November). Both events have led to floods in the region. When and why such events appear is still not clear. The sulphate and base cations peaks at Lugano in 2010 were the consequence of the volcanic eruption at Eyafjellajokull (Iceland) in April 2010.

Figure 2.2: Mean annual concentrations in wet deposition at the sampling sites. Base cations are defined as the sum of calcium, magnesium and potassium.
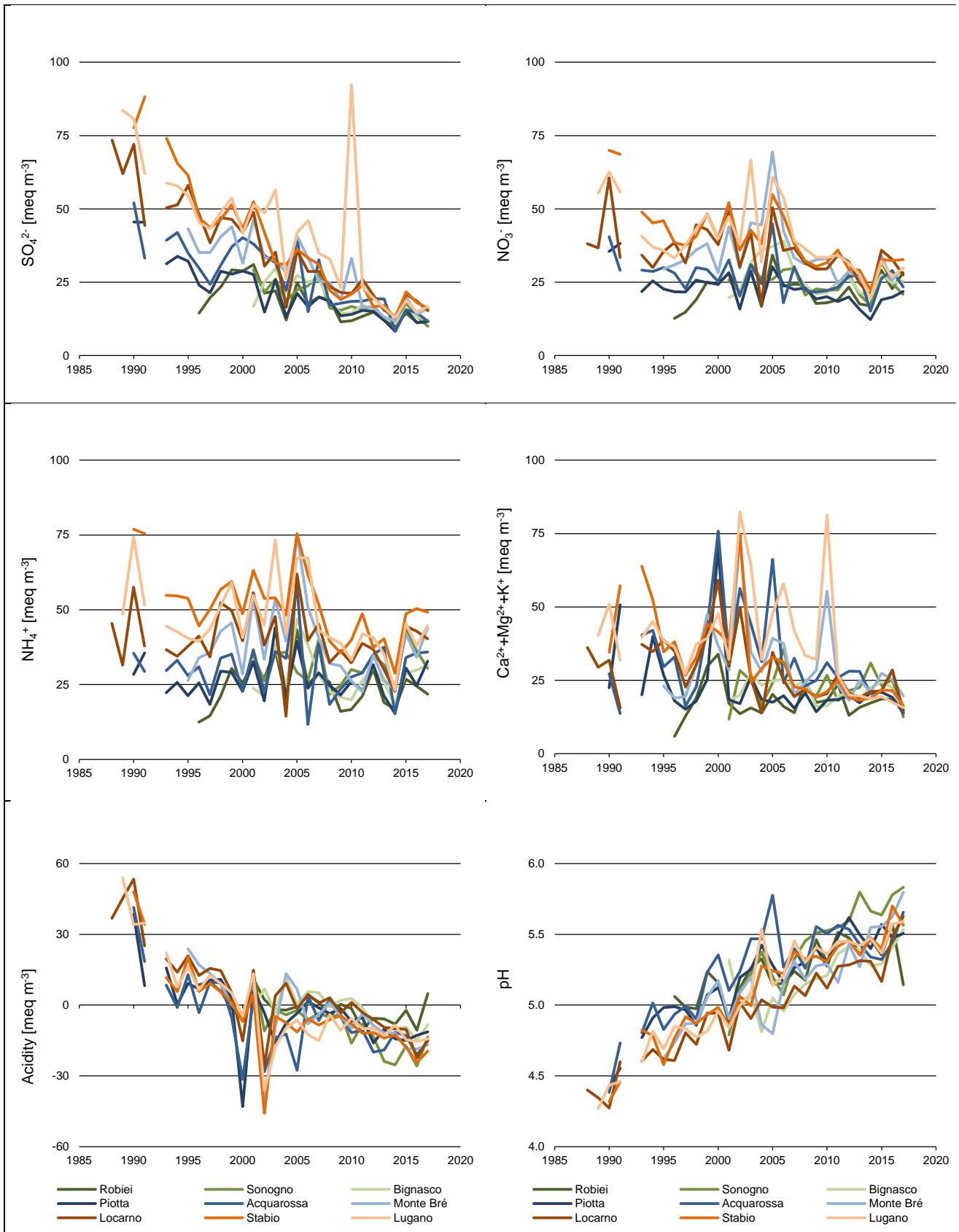
2.4 Trends in rainwater quality

2.4.1 Statistical methods

Trend analyses were performed on the key variables involved in acidification: sulphate, nitrate, ammonium, base cations (calcium, magnesium and potassium), H^+ and acidity. For each site and each parameter the mean monthly concentrations weighted with the precipitation volume were calculated and temporal trends were tested with the seasonal Mann-Kendall test (Hirsch et al. 1982) with a correction among blocks (Hirsch and 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).

2.4.2 Results from trend analysis

Trends of rainwater concentrations were analysed for two different time periods: from 1988-1991 until 2000 and from 2000 until 2017 (Tab. 2.3). Sulphate concentrations decreased at all sites and changes in concentrations were higher before 2000, except for Acquarossa. In contrast, nitrate and ammonium started to decrease significantly only after 2000 (7 out of 9 for nitrate and 4 out of 9 for ammonium). Before 2000 a significant decrease could only be observed at Stabio. Because of the decrease in sulphate and nitrate concentrations, concentrations of hydrogen ions and total acidity decreased significantly at all sites, although the changes in concentrations were higher before 2000. In general, concentrations of acidity decreased from values around 30-40 meq/m^3 to values around -15 meq/m^3 on average over the last 30 years. Accordingly, average pH increased from values around 4.3 in the 1990's to values ranging between 5.3 and 5.7 today.

Table 2.3 Changes in concentrations in rainwater expressed in meq m^{-3} yr^{-1} Significant trends are indicated in red.

	SO$_4$$^{2-}$		NO$_3$$^-$		NH$_4$$^+$		Cl$^-$		Base cations		H$^+$		Total acidity	
	'80/'90-00	'00-17	'90-00	'00-17	'90-00	'00-17	'90-00	'00-17	'90-00	'00-17	'90-00	'00-17	'90-00	'00-17
Acquarossa	-1.41	-1.52	-1.04	-0.41	-1.04	-0.17	-0.83	-0.04	-0.03	-1.86	-2.29	-0.02	-4.53	-0.11
Bignasco		-0.82		-0.58		-0.22		-0.02		-0.22		-0.49		-1.20
Monte Brè		-1.22		-0.64		-0.36		0.04		-0.45		-0.45		-1.68
Locarno Monti	-3.20	-1.45	-0.78	-1.05	-0.54	-0.68	-0.61	-0.05	-0.70	-0.57	-3.48	-0.59	-4.38	-1.40
Lugano	-2.79	-2.35	-1.22	-1.49	-0.10	-1.14	-0.70	-0.24	-0.63	-1.83	-2.85	-0.21	-4.28	-0.92
Piotta	-1.43	-0.67	-0.62	-0.53	-0.11	-0.30	-0.43	-0.05	-1.10	-0.31	-1.63	-0.25	-2.14	-0.59
Robiei		-0.76		-0.14		-0.44		0.00		-0.16		-0.21		-0.40
Sonogno		-0.63		-0.33		-0.04		0.06		-0.03		-0.27		-1.31
Stabio	-3.44	-1.70	-2.08	-0.96	-0.85	-0.75	-0.98	-0.05	-2.93	-0.63	-2.65	-0.23	-3.83	-1.27

2.5 Multiple regression analysis

In former reports it has been shown that the geographic distribution of the concentrations of sulphate, nitrate, ammonium and base cations in rainwater of Southern Switzerland can be described with a multiple linear regression model with the variables latitude, longitude and altitude (Barbieri and Pozzi 2001, Steingruber and Colombo 2010, Steingruber, 2015).

For this purpose for the 10 Swiss and 8 Italian sampling sites (Tab. 2.1), 5-years mean concentrations of sulphate, nitrate, ammonium and base cations weighted with the precipitation volume were calculated from yearly mean concentrations for the periods 1983-1987,1988-1992, 1993-1997, 1998-2002, 2003-2007, 2008-2012, 2013-2017. Since among geographically close sites yearly mean concentrations correlated significantly, missing annual mean concentrations were estimated from linear regression equations obtained plotting datasets of two sampling sites against each other. With this procedure for all 17 sampling sites (excluded the Basodino glacier) annual mean concentrations of all parameters could be reconstructed for the entire monitoring period 1983-2017. Missing mean winter concentrations of the Basodino glacier were estimated with the same procedure from mean winter concentrations measured and estimated at Robiei. To estimate annual mean concentrations at the Basodino glacier, 5-year mean winter concentrations were multiplied with annual mean/winter mean concentrations ratios at Robiei calculated for every 5-years period. Average mean concentrations for the different parameters and time periods are reported in Tab. A3 of the Appendix.

Multiple linear regression analyses were then performed for sulphate, nitrate, ammonium and base cations for the periods of 1983-1987,1988-1992, 1993-1997, 1998-2002 and 2003-2007, 2008-2012, 2013-2017. Parameters for the following multiple linear regressions were derived:

$C = m_{long}*longitude + m_{lat}*latitude + m_{alt}*altitude + C_0$

where:

C = mean concentration weighted with the amount of precipitation over the studied time period

C_0 = intercept

m_{lat}, m_{long}, m_{alt} = linear regression coefficients (=slopes)

Longitude, latitude and altitude are given in m (Swiss projection CH1903 LV03).

The linear regression coefficients for sulphate, nitrate, ammonium, base cations and the values describing the statistic significance of the regression model are reported in Tab. A4 of the Appendix. Concentrations of sulphate and nitrate depend always significantly on latitude and altitude. Concentrations of ammonium depend significantly always on latitude and only occasionally on altitude (1998-2002, 2003-2007, 2013-2017) probably because local emissions are less important. For describing the geographic distribution of the concentrations of base cations, longitude together with latitude are the most important parameters.

3. Wet deposition

3.1 Geographic interpolation

The multiple parameter regression model described in the previous chapter permitted the calculation of concentrations maps. The area under investigation was divided into 1km x 1km cells. For every cell center a concentration of the chemical parameter for the corresponding longitude, latitude and altitude was calculated.

Wet deposition maps of sulphate, nitrate, ammonium and base cations were obtained by multiplying concentration maps with precipitation maps.

3.2 Maps

Wet deposition maps of sulphate, nitrate, ammonium and base cations are shown in Fig. 3.1-3.4 . In general the geographic distribution is similar to that described for rainwater concentrations with sulphate, nitrate and ammonium decreasing along a south to north and an altitude gradient.

Wet deposition values also changed with time. A significant decrease in deposition of especially sulphate but also of nitrate and ammonium can be observed. Wet deposition of base cations also decreased slightly with time. Particularly rain rich and rain poor years can have visible consequences on deposition. As an example deposition of nitrate, ammonium and base cations were slightly higher during the rain rich 1998-2002 period compared to the immediately previous and successive time periods.

Figure 3.1: Wet deposition of sulphate
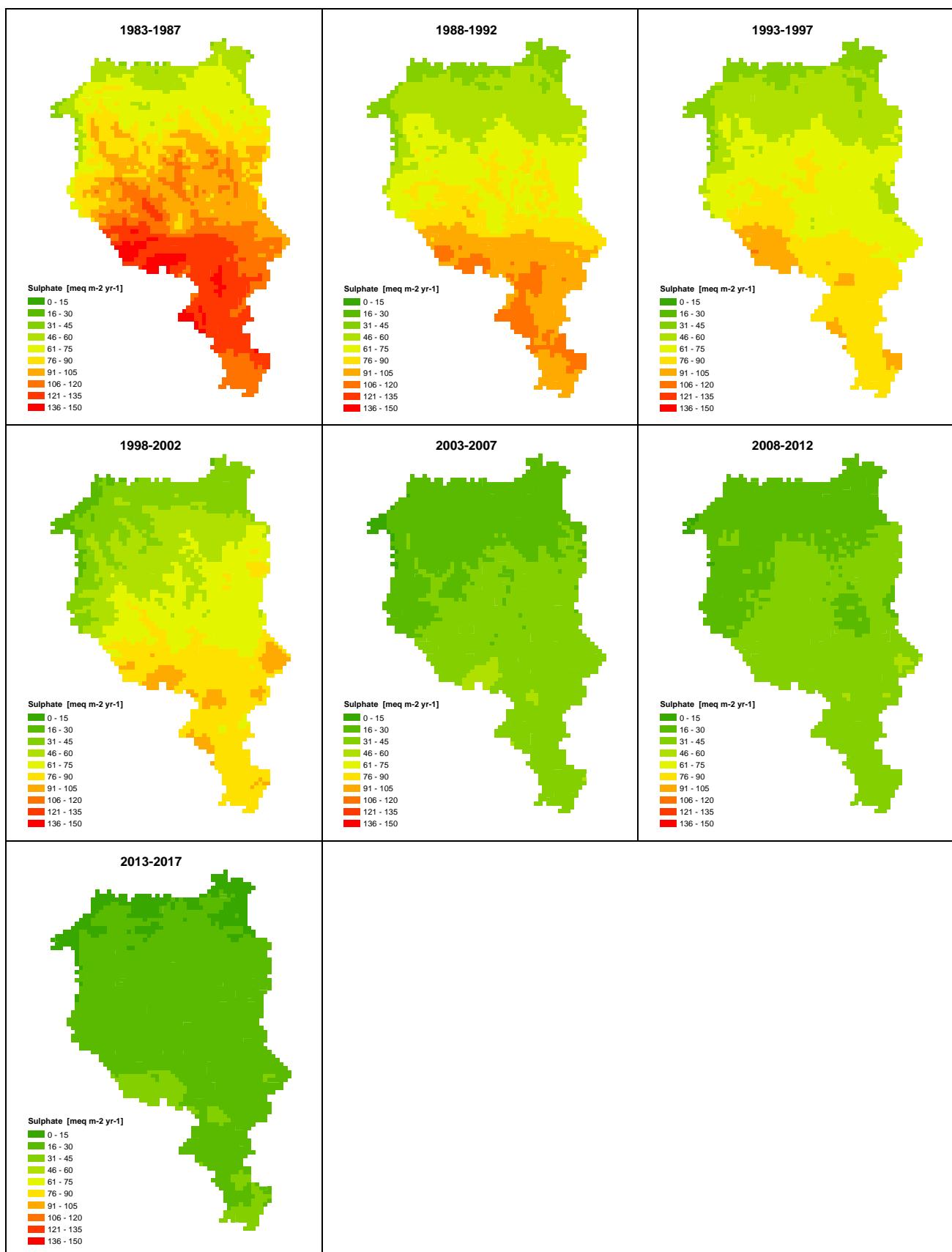
Figure 3.2: Wet deposition of nitrate
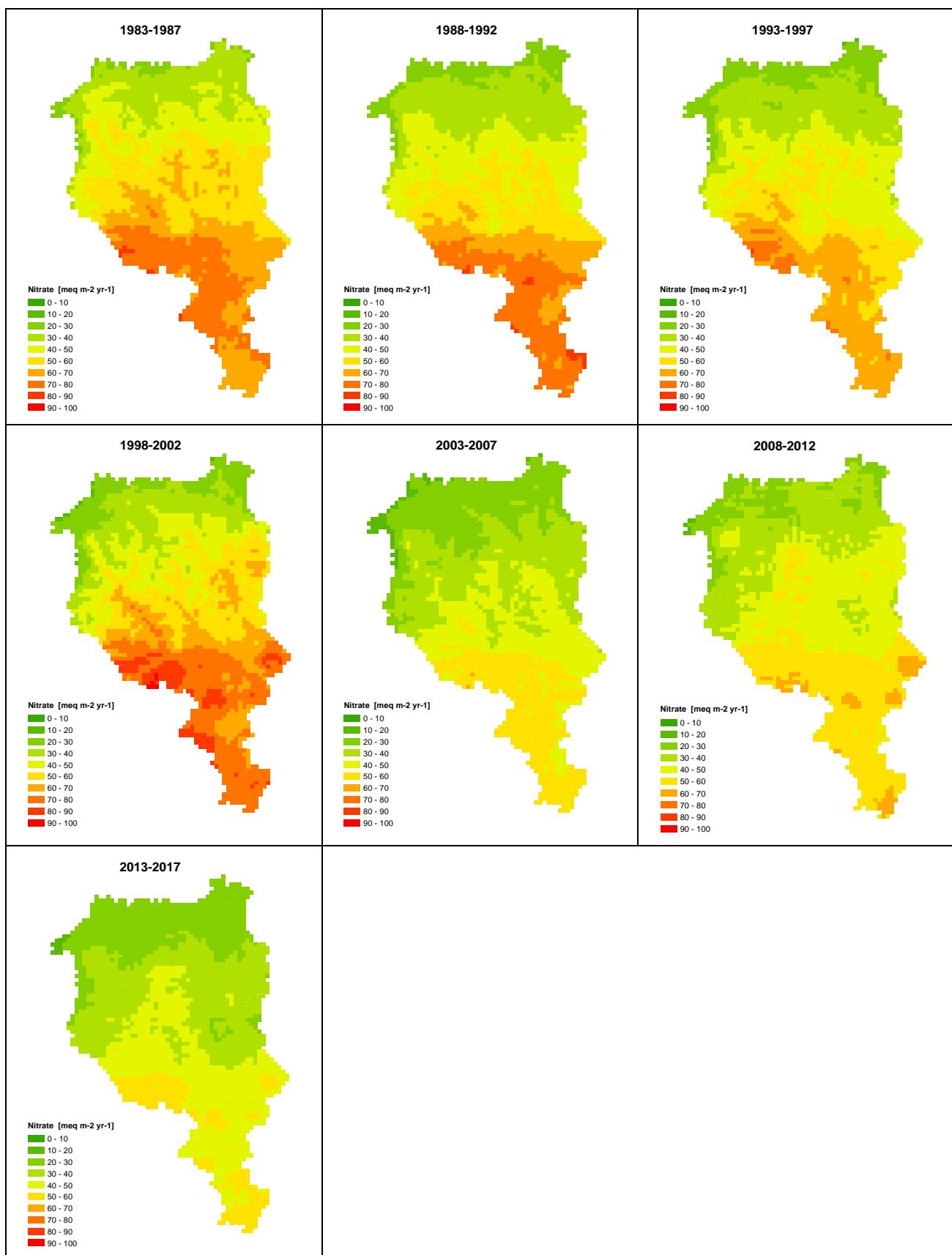
Figure 3.3: Wet deposition of ammonium
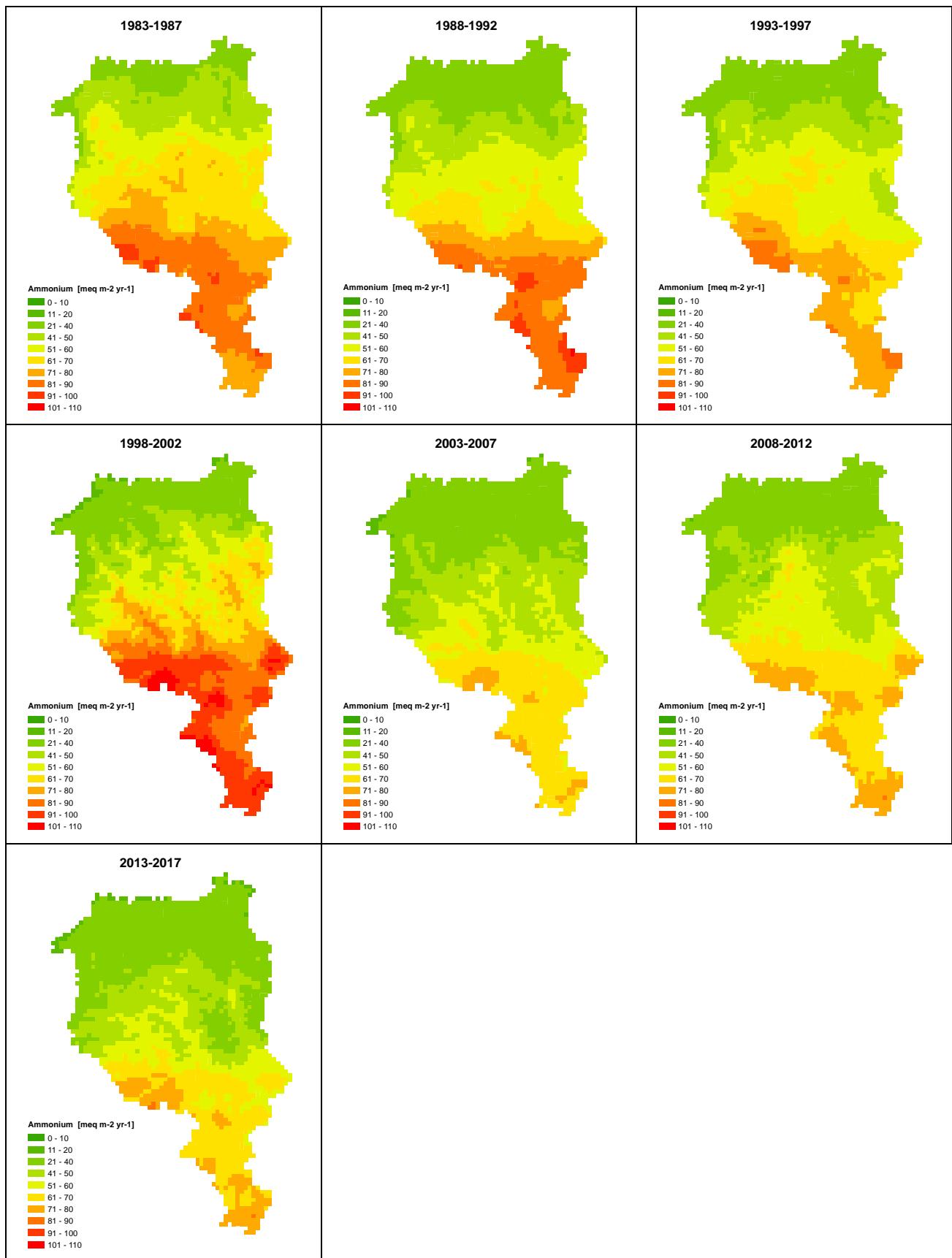
Figure 3.4: Wet deposition of base cations (Ca^{2+}+Mg^{2+}+K^+)
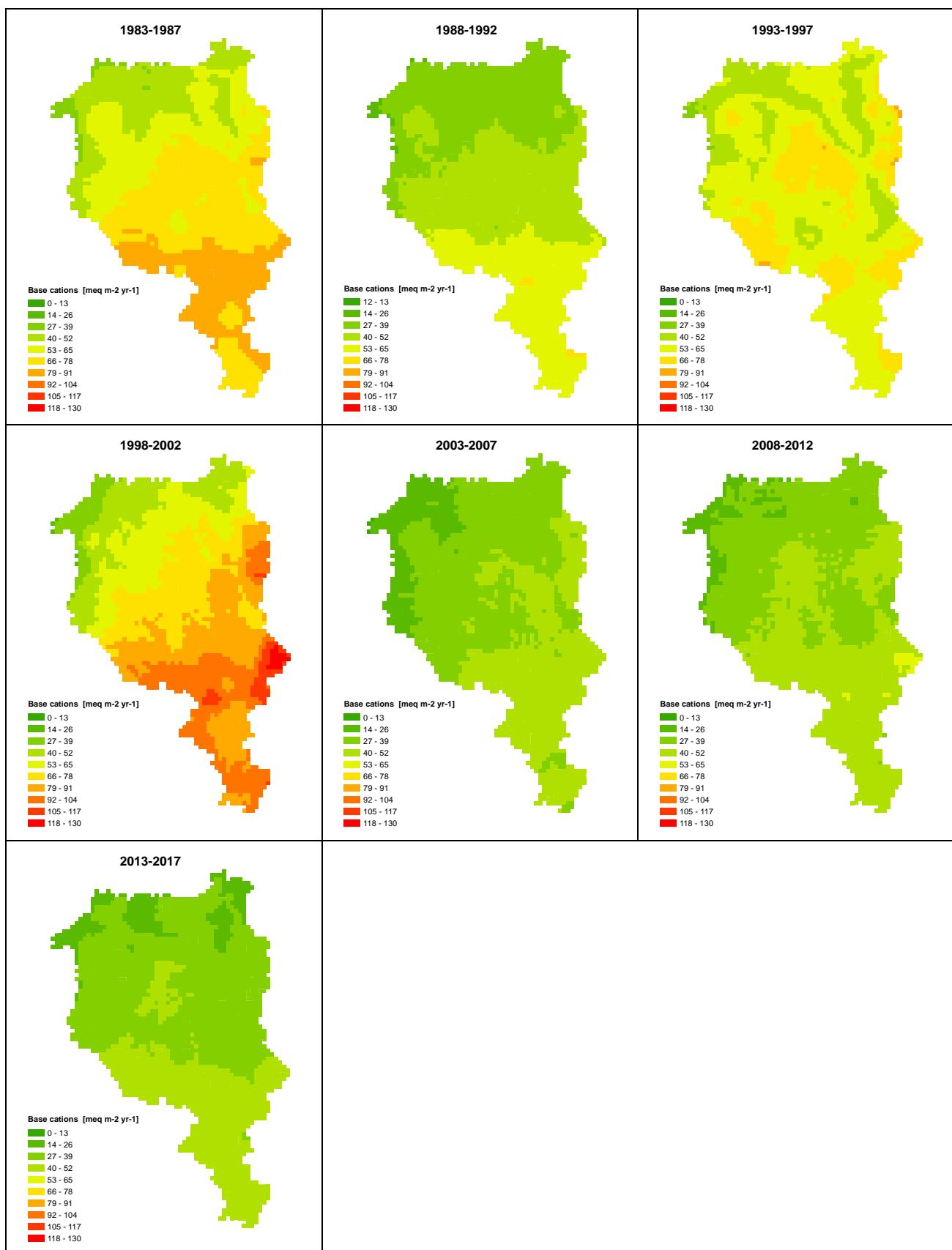
4.   Dry deposition

4.1   Mapping methods

Besides wet deposition, dry deposition of gases and aerosols also contribute to total deposition. For quantifying total acidifying deposition, dry deposition of the gaseous compounds NH_3, NO_2, SO_2 , HNO_3 and of the NH_4^+- and NO_3^--containing aerosols has to be known. Dry deposition of sulphate is not considered since its values are negligible compared with those due to wet deposition (Hertz and Bucher 1990). SO_2 and NO_x are emitted from combustion of fossil fuels, HNO_3 is formed by photochemical oxidation of NO_2, while NH_3 is mainly emitted from livestock breeding and from use of mineral fertilizers.

Unlike wet deposition, dry deposition cannot be measured directly. Therefore, yearly dry deposition maps are calculated by Meteotest on behalf of the Federal Office for the Environment multiplying modelled air concentrations (annual means, see https://www.bafu.admin.ch/bafu/de/home/themen/luft/zustand/daten/luftbelastung--historische-daten/karten-jahreswerte.html) with average deposition velocities (FOEN, 2016). For the present report Meteotest provided deposition maps for the periods 1988-1992, 1993-1997, 2003-2007, 2008-2012, 2013-2017. Due to lacking data dry depositions of the period 1998-2002 were calculated by averaging values of 1993-1997 and 2003-2007.

Since there is almost no measurement for dry depositions of non-marine base cations, values modelled by EMEP for the year 2000 were used for the calculations. Wet and dry deposition values of calcium, magnesium and potassium of the 3 main 50km x 50km grid falling in Canton 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.

4.2   Maps

Dry depositions of SO_2, oxidized and reduced nitrogen and base cations are mapped in Fig. 4.1-4.4. For all parameters and time periods depositions decreased with altitude. As a result of reduced emissions, dry deposition of SO_2 and oxidized nitrogen decreased during the last 30 years. Almost no change with time occurred for dry deposition of reduced nitrogen.

Figure 4.1: Deposition of sulphur dioxide
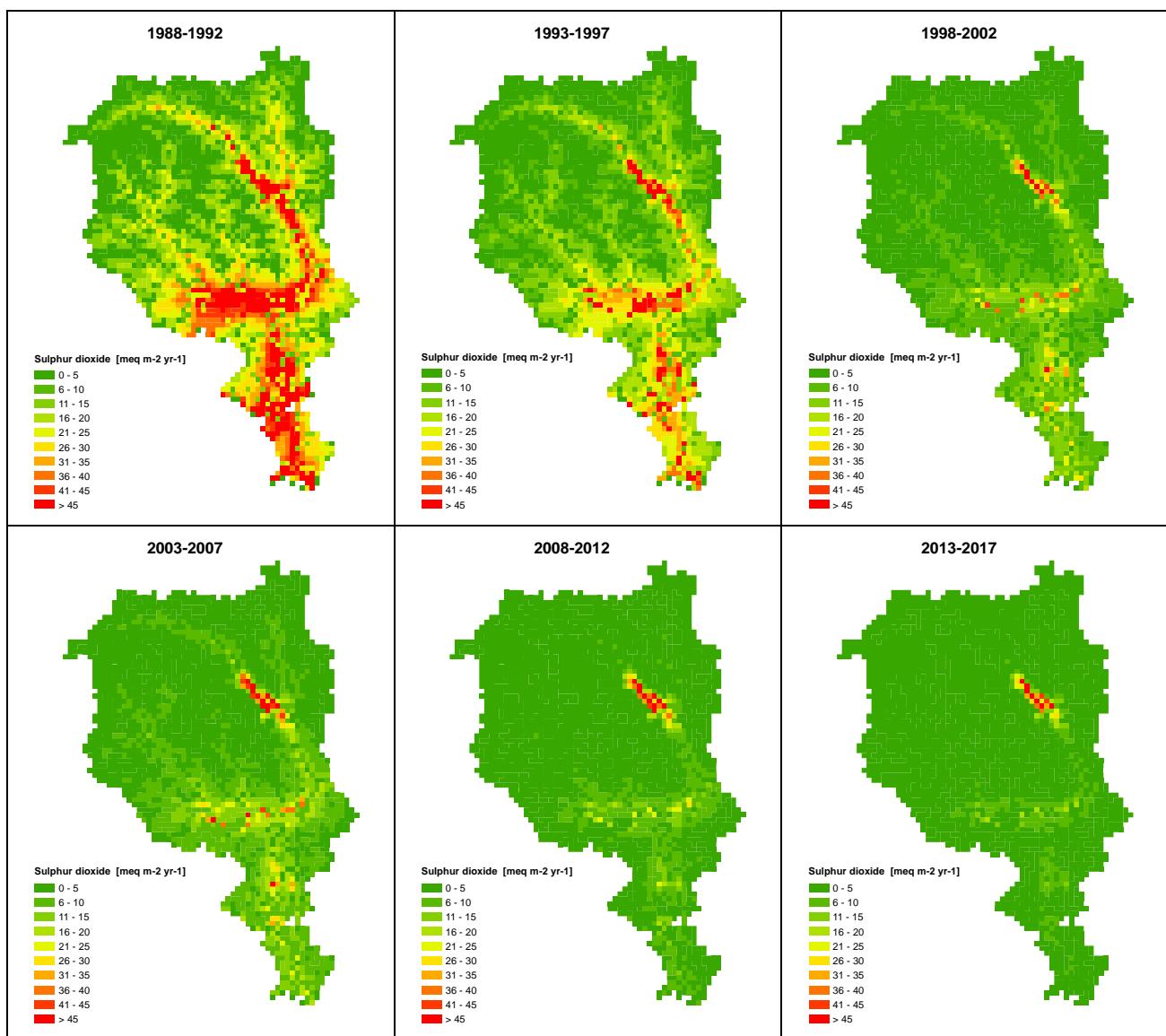
Figure 4.2: Dry deposition of oxidized nitrogen
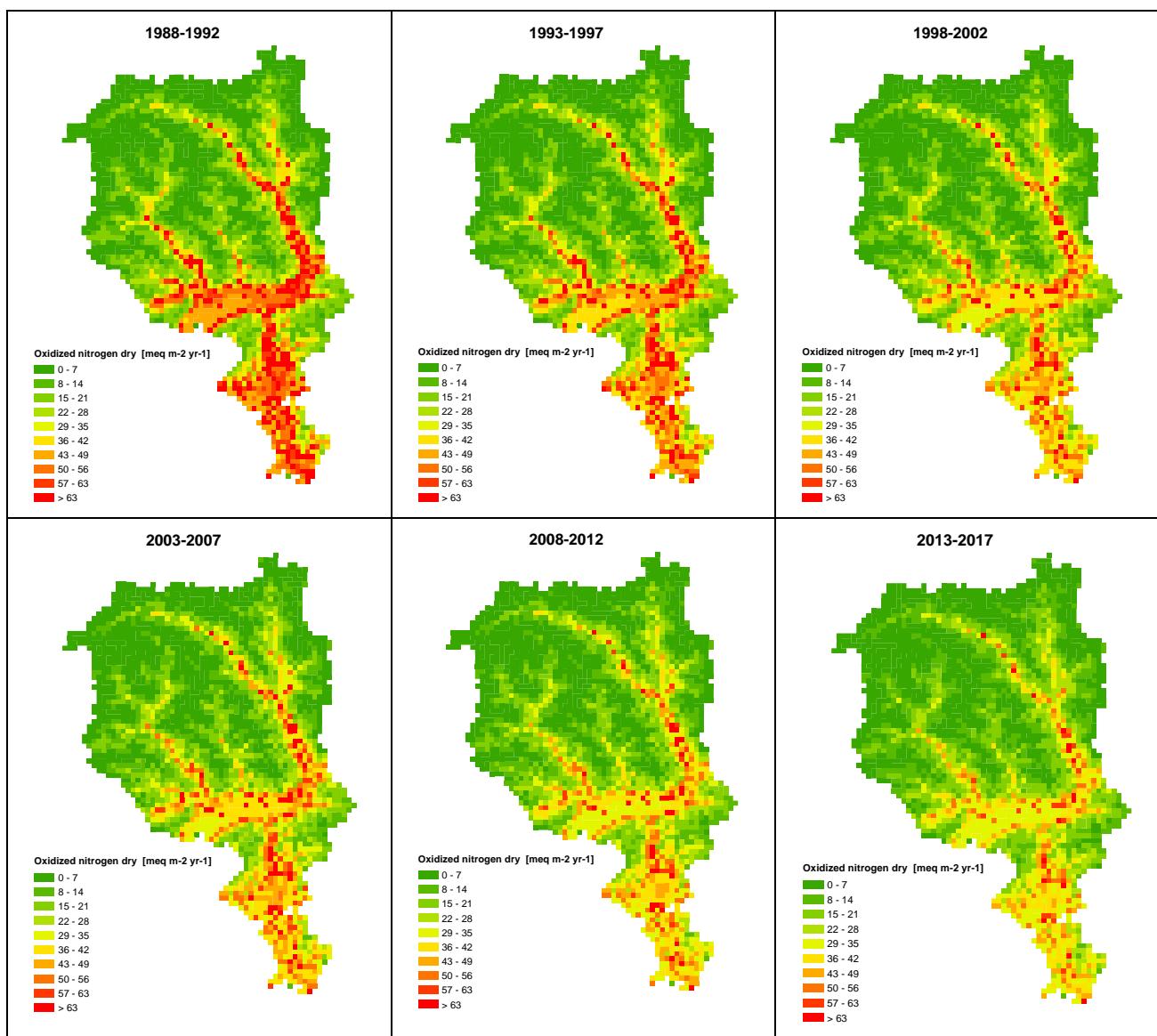
Figure 4.3: Dry deposition of reduced nitrogen
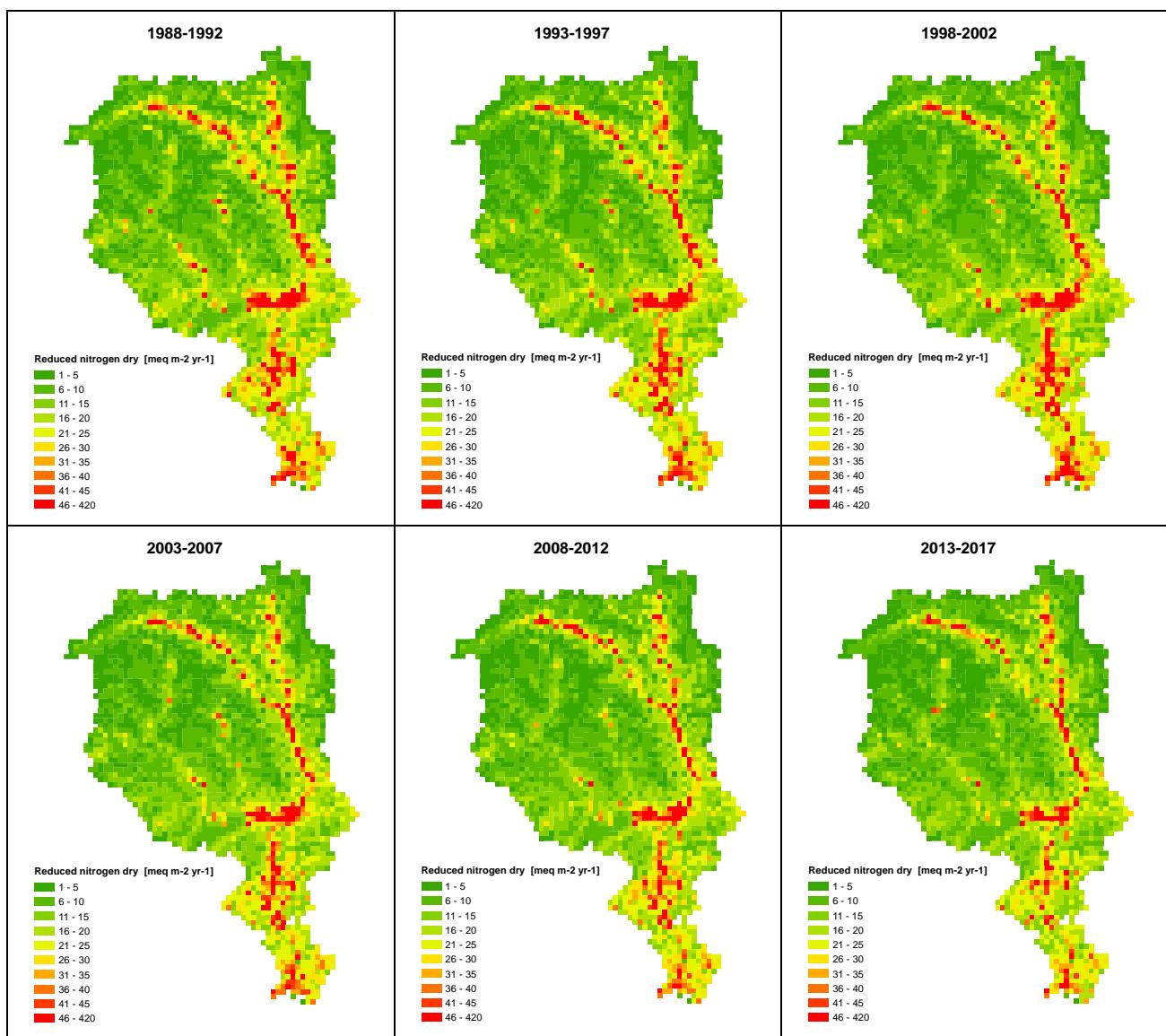
Figure 4.4: Dry deposition of base cations
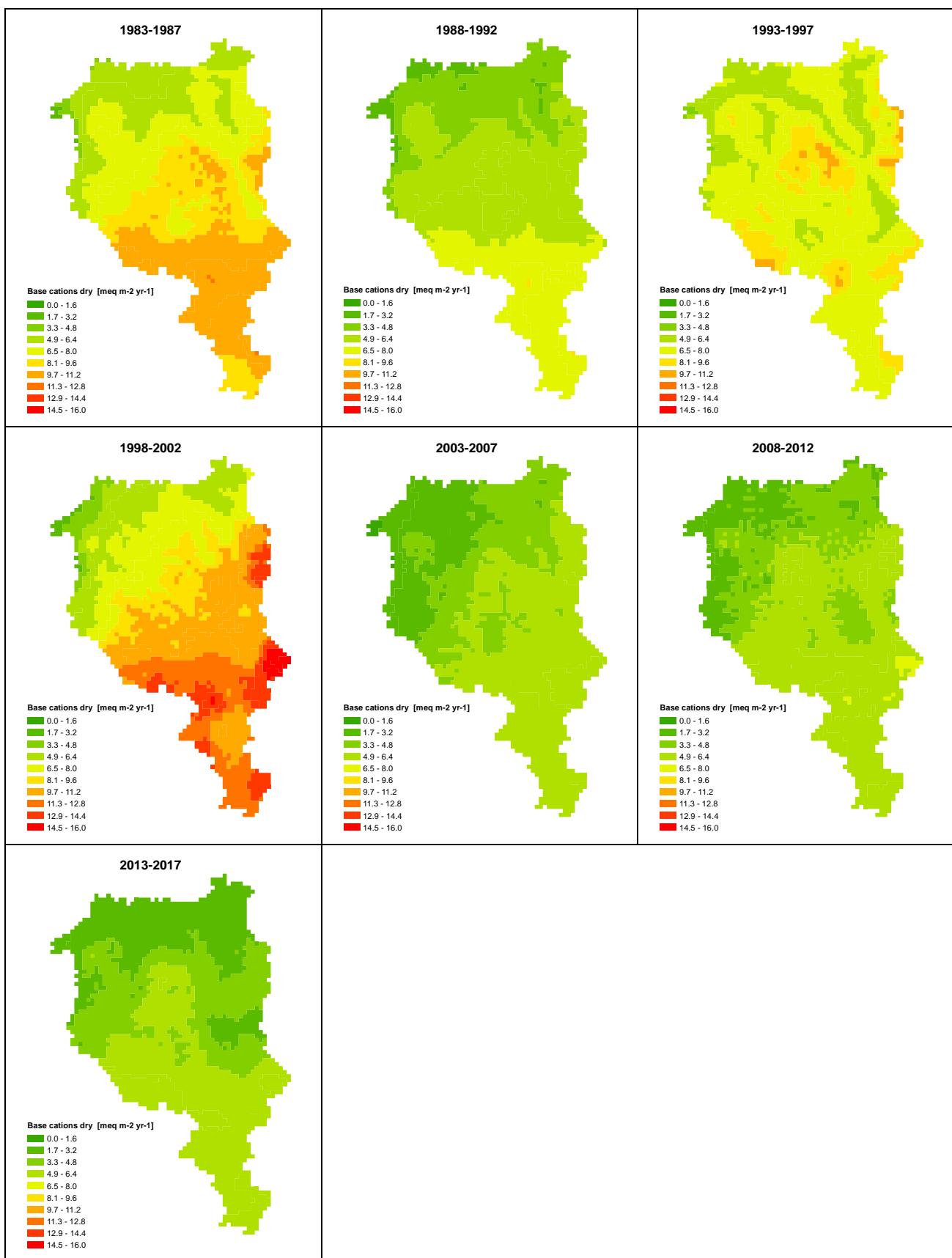
5. Total deposition

5.1 Mapping methods

Fig. 5.1-5.5 illustrate the maps for total deposition of sulphur, oxidized nitrogen, reduced nitrogen, total nitrogen, total base cations and the present load of acidity. The latter is also known as potential acidity since ammonia is considered as a potential acid.

These maps were produced by adding up the maps of wet and dry depositions discussed in the previous chapters. Since for the period 1983-1987 dry deposition maps were not available, maps 1988-1992 were used. The total deposition maps were then calculated as follows:

Total sulphur deposition:
wet deposition (SO_4^{2-}) + dry deposition (SO_2)

Total oxidized nitrogen deposition:
wet deposition (NO_3^-) + dry deposition ($NO_2 + NO_3^- + HNO_3$)

Total reduced nitrogen deposition:
wet deposition (NH_4^+) + dry deposition ($NH_3 + NH_4^+$)

Total nitrogen deposition:
Total oxidized N + Total reduced N

Total base cations deposition:
wet deposition (BC) + dry deposition (BC)

Present load of acidity (PLA):
Total nitrogen deposition + Total sulphur deposition – Total BC deposition

5.2 Maps

Depositions of total sulphur, total nitrogen and PLA decrease from south to north and from low to high altitude (Fig. 5.1, 5.4, 5.6). Total deposition of sulphur and nitrogen decreased consistently during the monitored period of time. Average total deposition of sulphur and nitrogen decreased from 114 to 25 meq m^{-2} yr^{-1} and from 158 to 117 meq m^{-2} yr^{-1}, respectively. Oxidized and reduced nitrogen contributed with about 50% each to the total. As a consequence of the reduction of sulphur and nitrogen deposition, deposition of the PLA also decreased significantly. Average PLA decreased from 202 to 104 meq m^{-2} yr^{-1}.

Tab. 5.1 presents the relative contribution of wet and dry sulphur and nitrogen deposition to the total acidifying load. Wet deposition contributes most to total deposition of acidifying compounds (between 71% and 79%), depending on the amount of yearly precipitation. Dry deposition is therefore less important. The contribution of sulphur compounds to total deposition of acidifying compounds decreased from 42% to 18%. This is explained by the stronger reduction of sulphur emissions over time compared to that of nitrogen. Accordingly, nitrogen compounds became more important in determining acidifying deposition. In fact, the percentage contribution to total acidifying deposition of reduced and

oxidized nitrogen compounds increased from 29% to 42% and from 29% to 40%, respectively.

Figure 5.1: Total deposition of sulphur
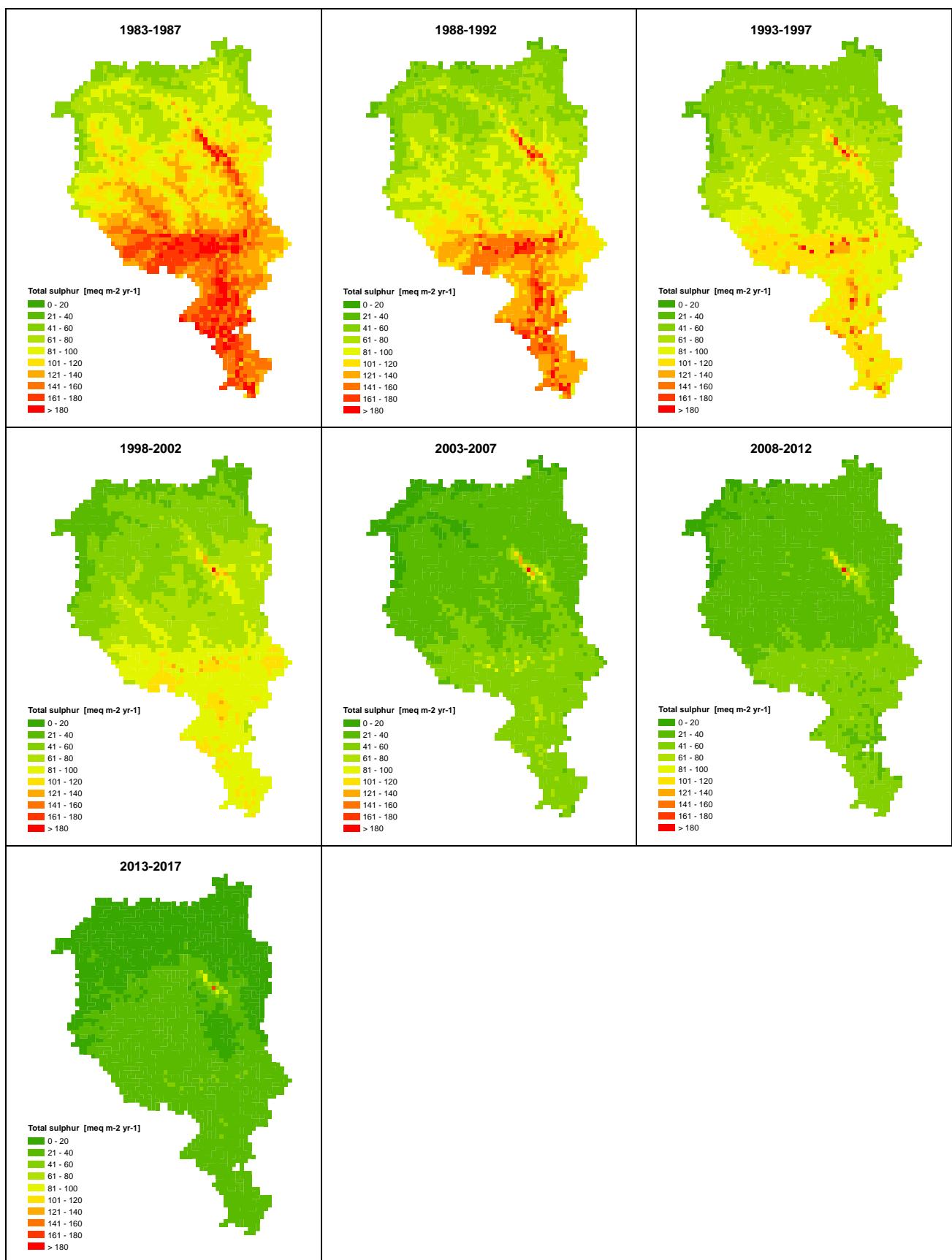
Figure 5.2: Total deposition of oxidized nitrogen
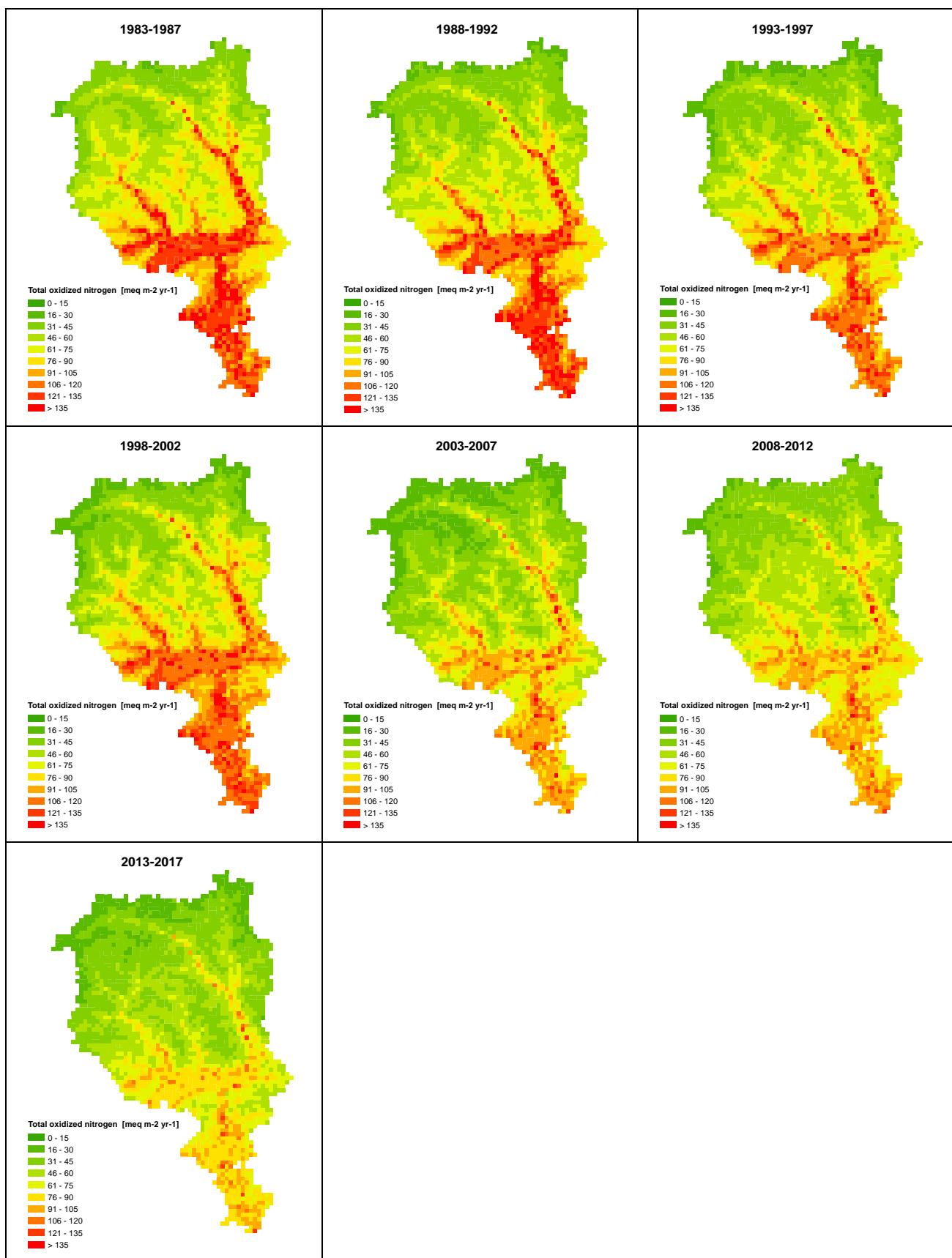
Figure 5.3: Total deposition of reduced nitrogen
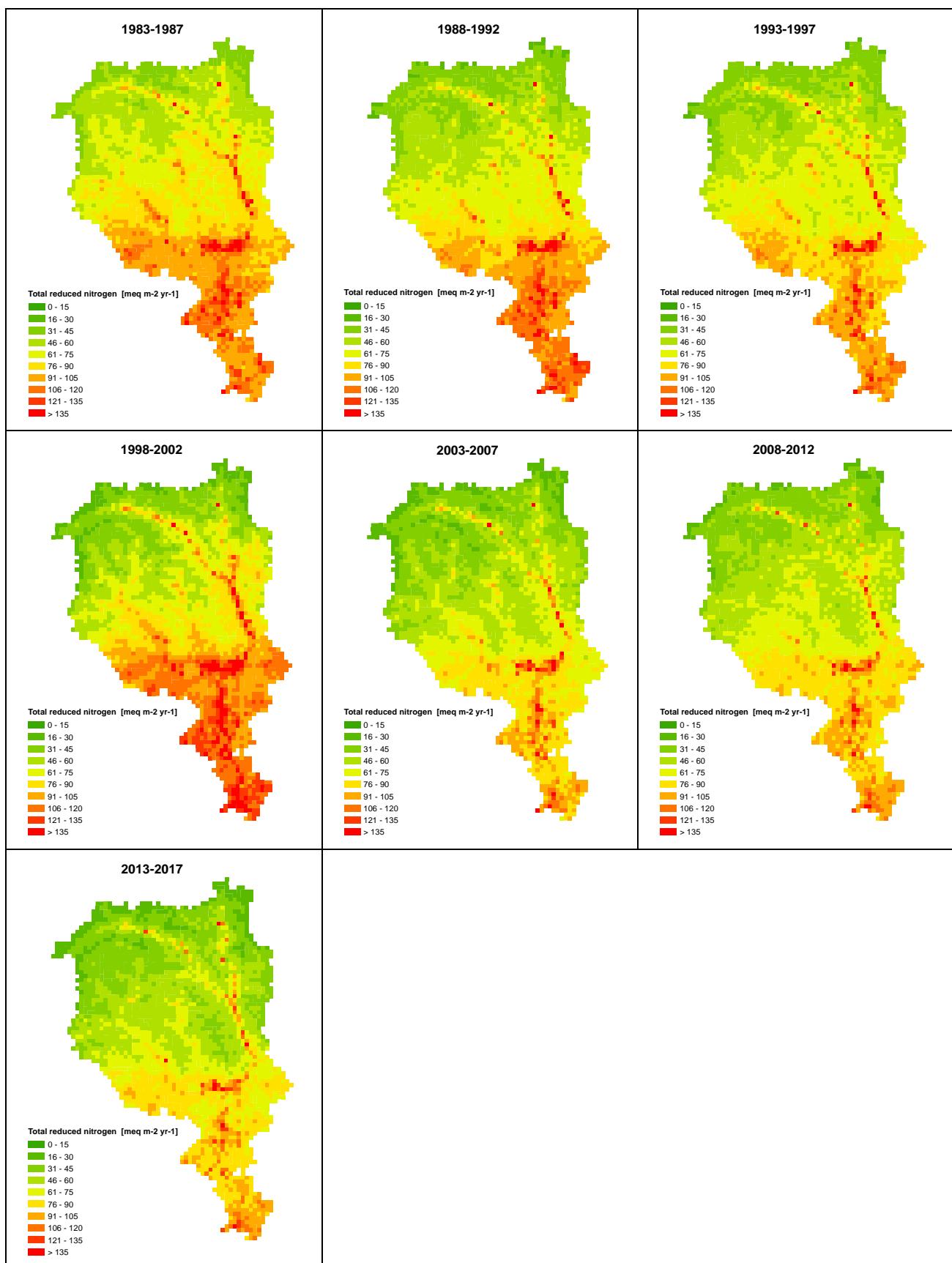
Figure 5.4: Total deposition of nitrogen
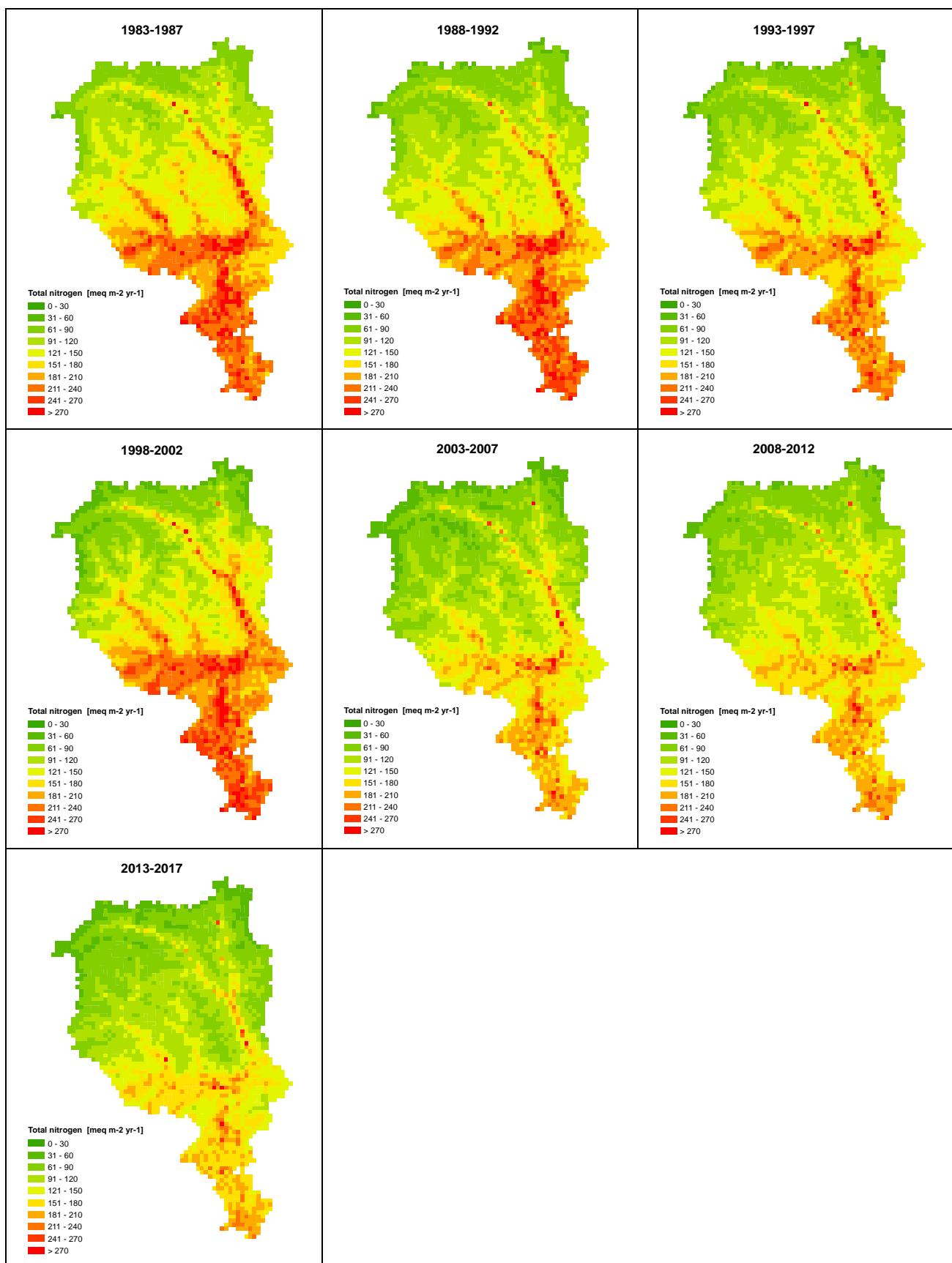
Figure 5.5: Total deposition of base cations
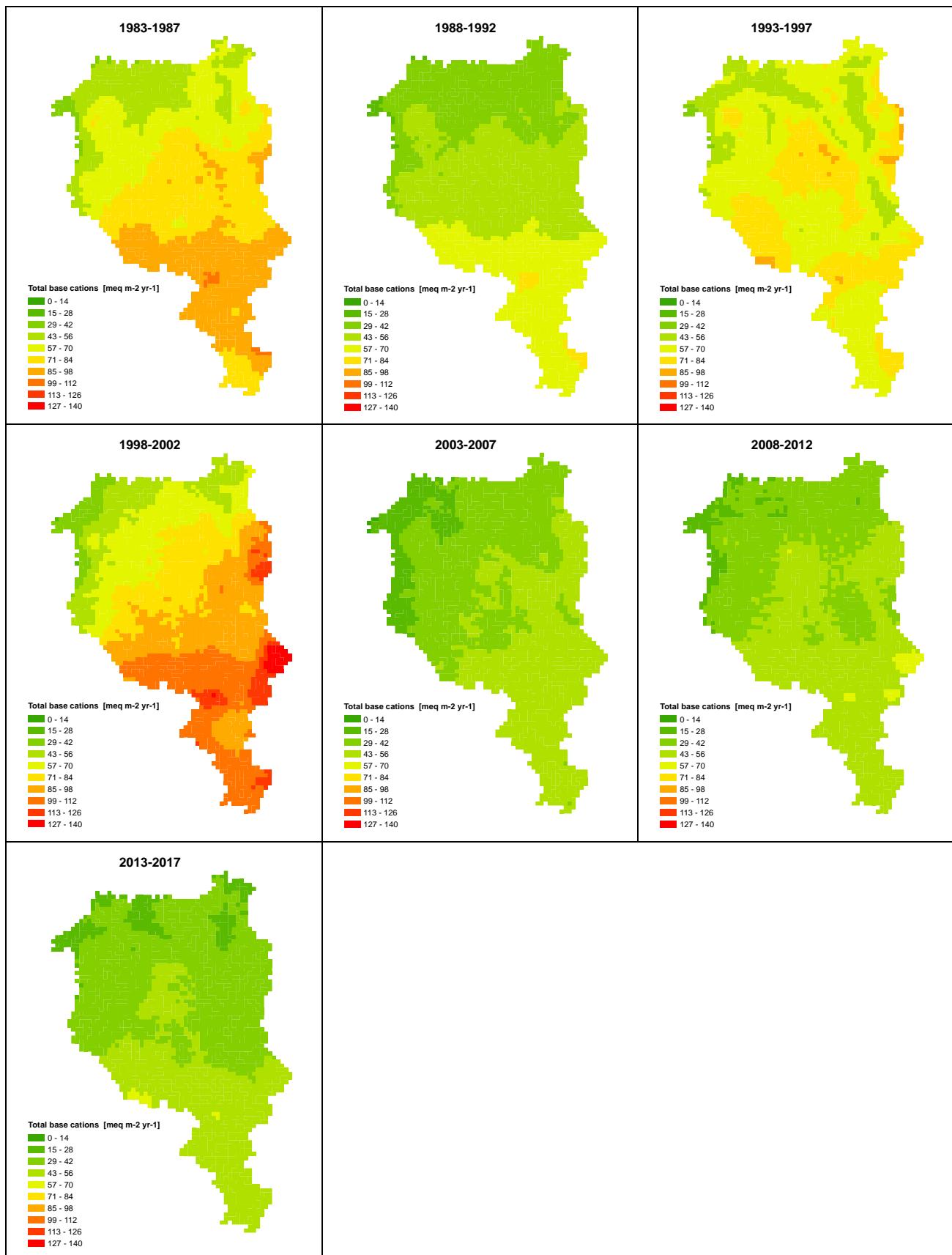
Figure 5.6: Deposition of potential acidity
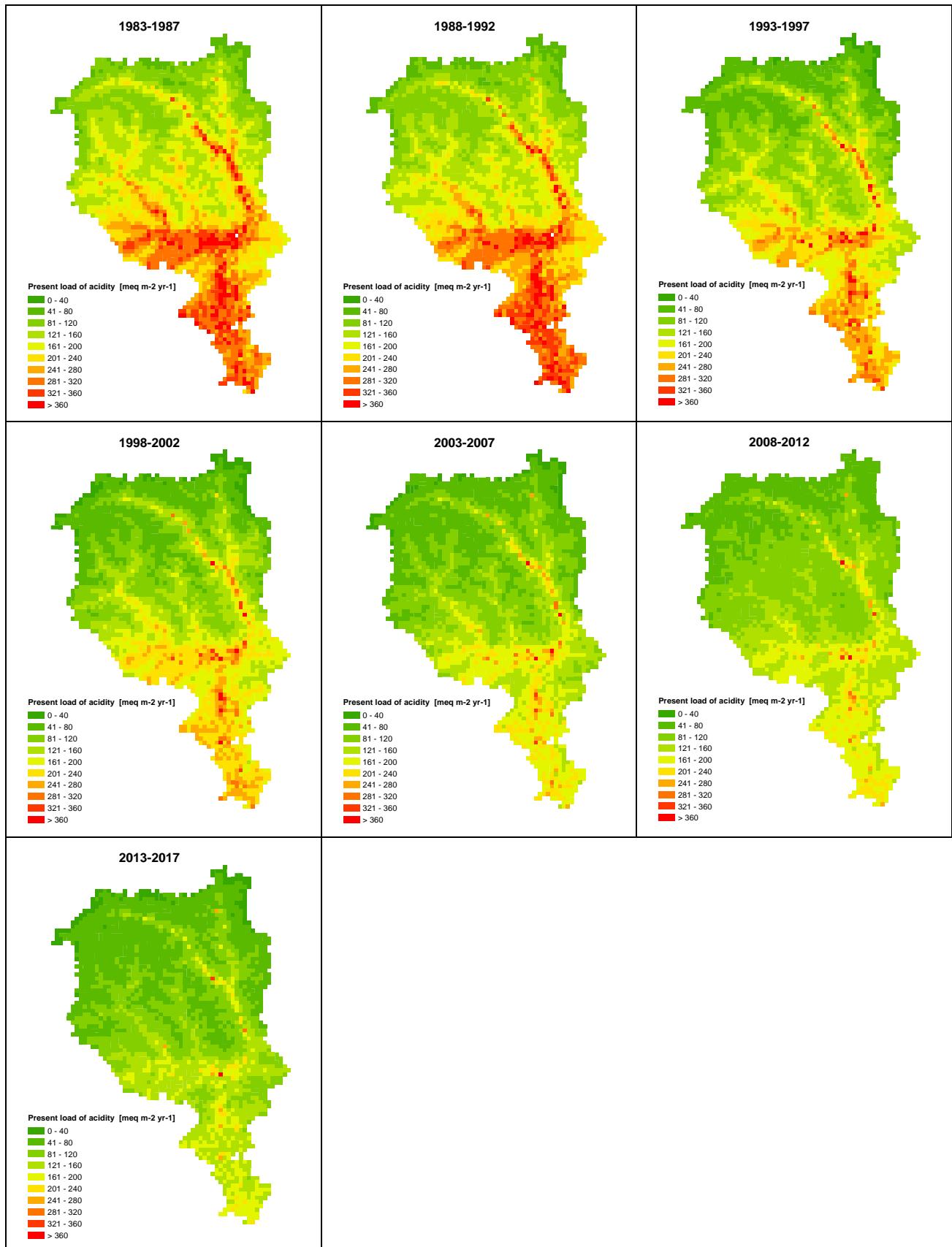
Table 5.1: Relative contribution of wet and dry nitrogen and sulphur deposition to total acidifying load

Period	Oxidized sulphur		Oxidized nitrogen		Reduced nitrogen	
	wet	dry	wet	dry	wet	dry
1983-1987	34%	8%	19%	10%	22%	6%
1988-1992	29%	9%	20%	11%	23%	7%
1993-1997	29%	7%	21%	11%	24%	8%
1998-2002	28%	3%	24%	10%	27%	8%
2003-2007	19%	5%	24%	14%	28%	10%
2008-2012	19%	3%	26%	13%	30%	10%
2013-2017	15%	3%	26%	14%	32%	11%

6. Exceedances of critical loads of acidity and nutrients at forest sites

6.1 Mapping methods

The temporal evolution of the exceedances of critical loads of nutrient nitrogen (CLnutN) and acidity (CLA) are shown here at the example of forest ecosystems. Switzerland regularly submits critical loads for different ecosystems to the Coordination Centre of Effetcs in the framework of the UNECE Convention on Long-range Transboundary Air Pollution (FOEN 2017).

For forest ecosystems critical loads of nutrient nitrogen (CLnutN) were modelled with the simple balance model (SMB; FOEN 2016, FOEN 2017). For Southern Switzerland CLnutN of 10 kg N ha^{-1} yr^{-1} (= 71 meq m^{-2} yr^{-1}) were submitted. A variant of the SMB model was used to derive a piece-wise linear critical load function for acidifying nitrogen and sulphur (FOEN 2017). Maps showing the exceedances of CLnutN at forest sites were calculated subtracting CLnutN from deposition of total nitrogen. Exceedances of critical loads of acidity were calculated by adding the N and S deposition reductions needed to reach the critical load function via the shortest path (Chapter 7; UNECE 2017).

6.2 Maps of exceedances

Maps of exceedances of CLnutN and CLA at forest sites are shown in Fig. 6.1 and Fig. 6.2, respectively. Between 1985 and 2015 average exceedance of CLA decreased from 134 to 30 meq m^{-2} yr^{-1} and the percentage of sites with exceeded CLA decreased from 81% to 26%. Average exceedance of CLnutN decreased (from 113 to 66 meq m^{-2} yr^{-1}). However, the percentage of sites with exceeded CLnutN remained unchanged and almost at all sites the CLnutN is still exceeded (99%).

Figure 6.1: Exceedance of CLNut at forest sites of the National Forest Inventory calculated
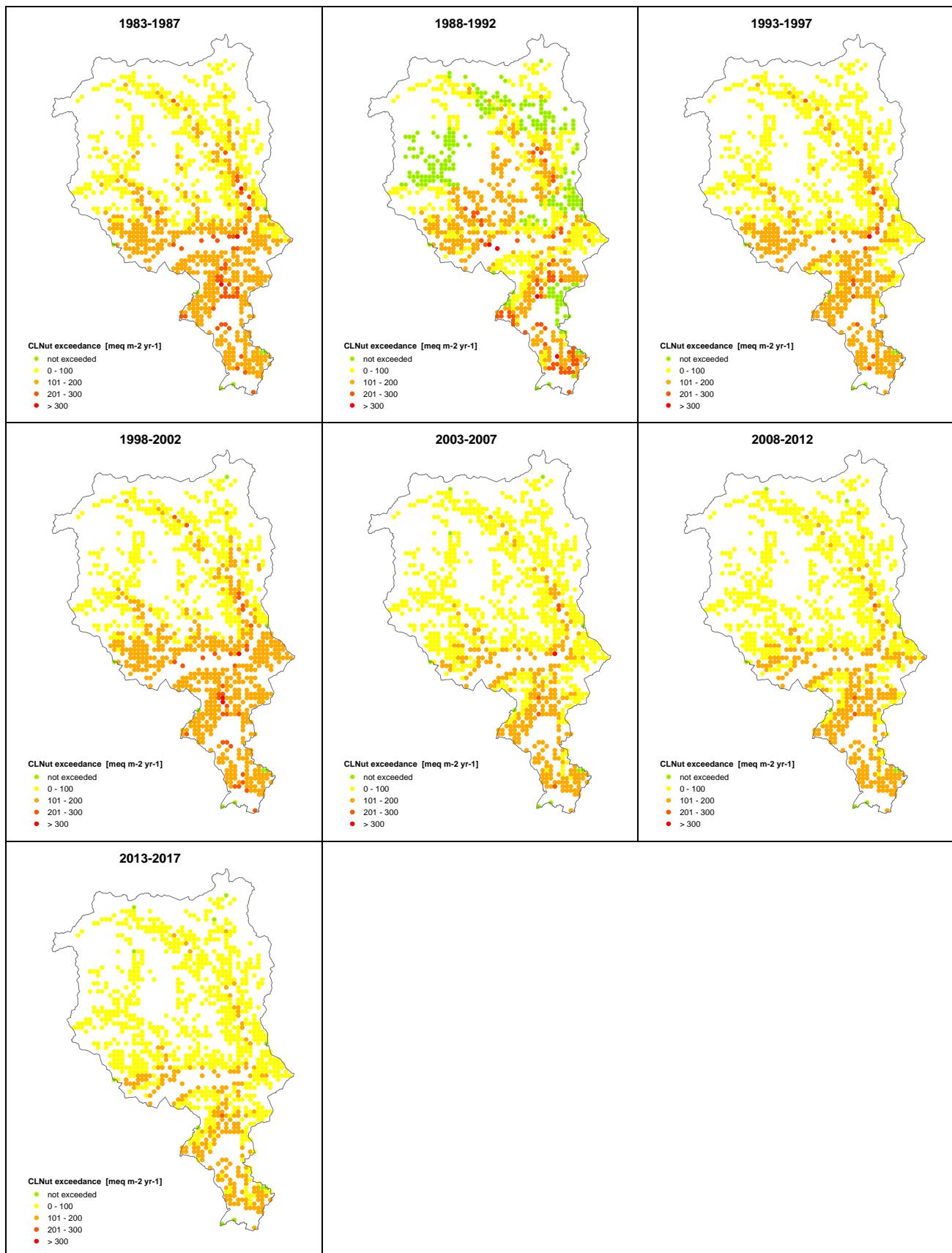
Figure 6.2: Exceedance of CLA at forest sites of the National Forest Inventory calculated as ΔS+ ΔN
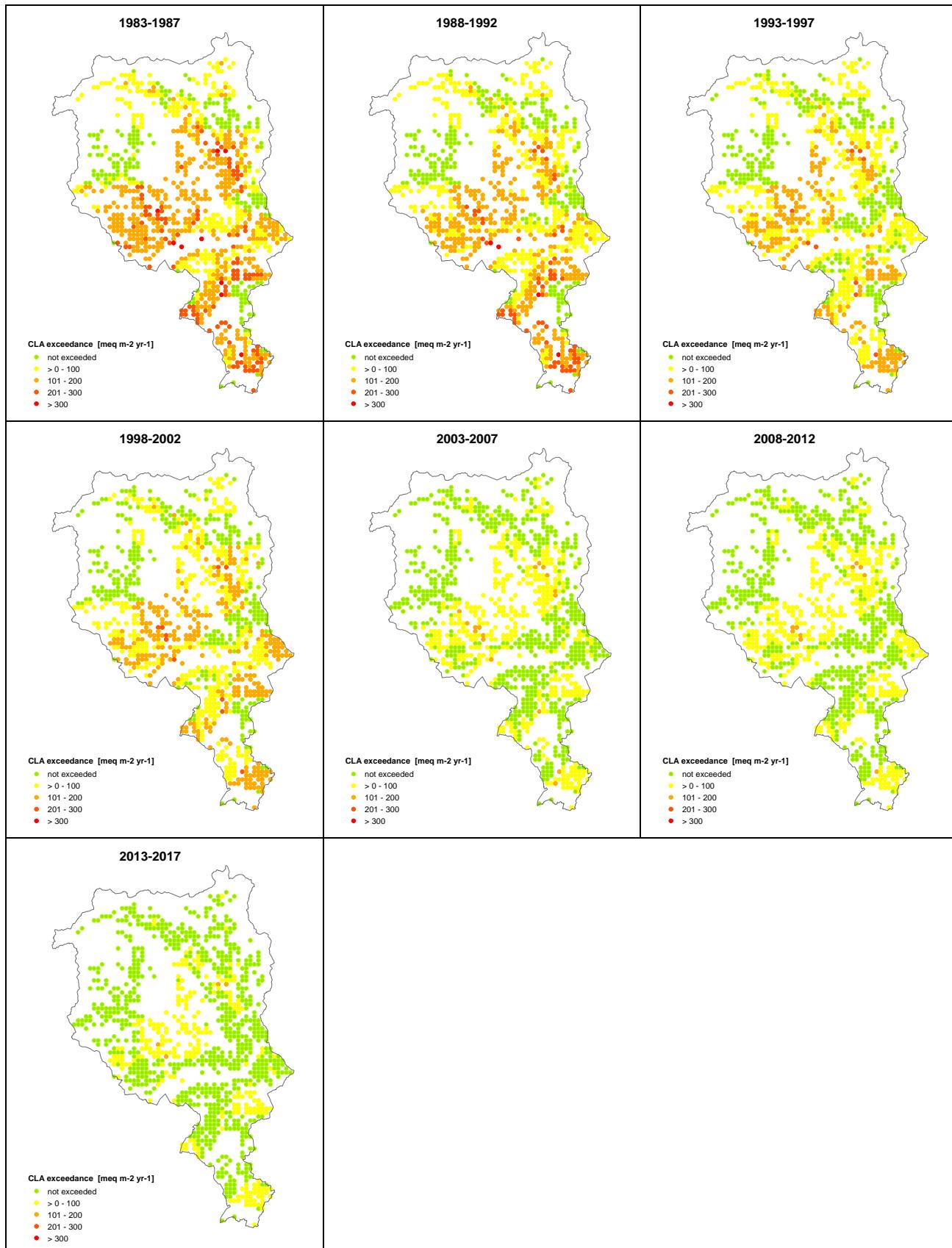
References

Austnes et al. 2018. Regional assessment of the current extent of acidification of surface waters in Europe and North America. NIVA report 7268-2018. ICP Waters Report 135/2018. Norwegian Institute for Water Research, Oslo, 134 p.

Barbieri A. and S. Pozzi. 2001. Acidifying deposition - Southern Switzerland. Environmental documentation No. 134. Swiss Agency for the Environment, Forests and Landscape (SAEFL, Ed.), Berne, 113 p.

Dillon P.J., N.D. Yan and H.H. Harvey. 1984. Acidic deposition. Effects on aquatic ecosystems. CRC Crit. Rev. Environ. Control 13: 167-194.

FOEN, 2016. Critical Loads of Nitrogen and their Exceedance – Swiss contribution to the effects-oriented work under the Convention on Long-range Transboundary Air Pollution (UNECE). Environmental studies no. 1642. Federal Office for the Environment (FOEN, Ed.), Berne, 78 p.

FOEN, 2017 National Focal Centre Report – Switzerland. In: Hettelingh J.-P., Posch M., Slootweg J. (eds), CCE Status Report 2017, Coordination Centre for Effetcs, RIVM, Bilthoven, pp. 177-190.

FOWG. 2000. Daily Precipitation maps 1961-1999, based on maps of the Hydrological Atlas of Switzerland and monitoring data from MeteoSwiss, Zürich. Federal Office for Water and Geology (FOWG, Ed.), Berne.

Heldstab J., Schäppi B., Weber F., Sommerhalder M. 2018. Switzerland's Informative Inventory Report 2018. Submission under the UNCECE Convention on Long-range Transboundary Air Pollution. Federal Office for the Environment, Berne, 345 p.

Hertz J. and P. Bucher. 1990. Abschätzung der totalen Stickstoff- und Protoneneinträge in ausgewählte Ökosysteme der Schweiz. VDI-Berichte 837: 373-387.

Hirsch R.M. and J.R. Slack. 1984. A nonparametric test for seasonal data with serial dependance. Water Resources Research 20: 727-732.

Hirsch R.M., J.R. Slack and R.A. Smith. 1982. Techniques of trends analysis for monthly water quality data. Wat. Res. Res. 18(1): 107-121.

ICP Waters Programme Centre. 2010. ICP Waters Progamme Manual 2010. NIVA report SNO. 6074-2010. ICP Waters Report 105/2010. Norwegian Institute for Water Research, Oslo, 91 p.

Künzler P. 2005. Weiterentwicklung des Luftreinhalte-Konzepts - Stand, Handlungsbedarf, mögliche Massnahmen. Schriftenreihe Umwelt Nr. 379. Bundesamt für Umwelt, Wald und Landschaft (BUWAL, Ed.), Bern, 171 p.

Odén S. 1968. The acidification of air and precipitation and its consequences on the natural environment. Ecology Committee, Bulletin No. 1. Swedish National Science Research Council, Stockholm, 117 p.

Marchetto A. 2014. rkt: Mann-Kendall test, Seasonal and Regional Kendall Tests. (ultimo aggiornamento 22.1.2014).

MeteoSvizzera. 2012. Rapporto sul clima – Cantone Ticino 2012. Rapporto di lavoro MeteoSvizzera no. 239, Ufficio federale di meteorologia MeteoSvizzera, Locarno Monti, 63 p.

EMEP. 2016. Transboundary particulate matter, photo-oxidants, acidifying and eutrophying components. Status Report 2016. Meteorological Synthesizing Centre - West, Norwegian Meteorological Institute, Oslo.

Rogora M., Colombo L., Lepori F., Marchetto A., Steingruber S. and Tornimbeni O. 2013. Thirty Years of Chemical Changes in Alpine Acid-Sensitive Lakes in the Alps. Water Air Soil. Pollut. 224: 1746.

Smith R.A. 1852. On the air and rain of Manchester. Memoirs of the Manchester Literary and Philosophical Society 10: 207-217.

Spinedi F. and F. Isotta. 2004. Il clima del Ticino. Dati - statistiche e società 2: 4-39.

Steingruber, S.M. 2015. Acidifying deposition in southern Switzerland – Monitoring, maps and trends 1983-2013. Dipartimento del territorio del Canton Ticino, Bellinzona, Switzerland, 60 p.

Steingruber, S.M. and L. Colombo. 2010. Acidifying deposition in southern Switzerland - Assessment of the trend 1988-2007. Environmental studies no. 1015, Federal Office for the Environment, Bern, 82 p.

UNECE, 2017. Manual on methodologies and criteria for Modelling and Mapping Critical Loads & Levels and Air Pollution Effects, Risks and Trends. Convention on Long-range Transboundary Air Pollution (UNECE). Distributed and updated at the joint Session of the Steering Body to the EMEP and the Working Group on Effects in September 2016.

Acknowledgments

Appendix

Table A1: Swiss (CH) and Italian (I) precipitation sampling sites and their Swiss coordinates (CH1903 LV03), altitudes, data source and period used for the calculation of depositions

N°	Sampling site	Longitude (m)	Latitude (m)	Altitude (m a.s.l.)	Date source	Period
1	Acquarossa/Comprovasco (CH)	714998	146440	575	MeteoSwiss	1983-2017
2	Agrasina (I)	674587	119675	1370	Idroelettriche Riunite S.p.A.	1983-2017
3	Airolo (CH)	688910	153400	1138	MeteoSwiss	1983-2017
4	Andermatt (CH)	687444	165044	1438	MeteoSwiss	1983-2017
5	Arcisate (I)	712824	78131	383	ARPA Lombardia	2008-2017
6	Arosio (CH)	713130	100610	860	UCA	1988-2017
7	Bedretto (CH)	682303	151023	1397	UCA	2008-2017
8	Bellinzona (CH)	720913	116588	224	MeteoSwiss	2008-2017
9	Biasca (CH)	718550	132800	278	MeteoSwiss	1983-2017
10	Biasca (CH)	717000	135125	293	UCA	2013-2012
11	Biasca Pontirone (CH)	723860	137864	1405	UCA	2008-2017
12	Bioggio (CH)	714170	96525	285	OASI	2008-2017
13	Bosco Gurin (CH)	680879	130027	1486	MeteoSwiss	1983-2017
14	Braggio (CH)	729975	128600	1315	MeteoSwiss	1983-2017
15	Brissago (CH)	698200	108390	280	MeteoSwiss	1983-2017
16	Bruggi (I)	676295	133657	1226	ARPA Piemonte	2013-2017
17	Bruzella (CH)	724090	82490	620	MeteoSwiss	1983-2007
18	Cabbio (CH)	724643	84028	1790	UCA	2013-2017
19	Cadero Veddasca (I)	703187	101753	570		1983-1997
20	Cala-Chironico (CH)	705195	141435	1460	UCA	2013-2017
21	Camedo (CH)	690296	112207	590	MeteoSwiss	1983-2017
22	Camedo (CH)	690050	112110	558	UCA	2003-2017
23	Camedo diga (CH)	690940	112220	520	UCA	1983-2007
24	Camignolo (CH)	715430	106905	435	OASI	2008-2017
25	Campo Vallemaggia (CH)	681711	126785	1303	UCA	2003-2017
26	Canobbio-Trevano (I)	717900	98520	342	UCA	2008-2017
27	Carena (CH)	727230	114230	942	UCA	2008-2017
28	Cavargna (I)	729353	105751	1100	ARPA Lombardia	2008-2017
29	Cavergno (CH)	690081	133073	455	UCA	2008-2017
30	Cevio (CH)	689688	130565	417	MeteoSwiss	1983-2017
31	Chiasso (CH)	722690	77090	240	UCA	1988-2017
32	Cimetta (CH)	704433	117452	1661	MeteoSwiss	1983-2017
33	Coldrerio (CH)	721080	79235	347	MeteoSwiss	1988-2012
34	Colla (CH)	725030	106400	1140	UCA	2008-2017
35	Copera (CH)	720318	112564	665	WSL	1983-1992
36	Crana-Torricella (CH)	712695	103746	1002	MeteoSwiss	1983-2017
37	Creva (I)	702820	94488	233		1983-1997
38	Disentis/Sedrun (CH)	708189	173789	1197	MeteoSwiss	1983-2017
39	Faido (CH)	704950	148266	747	MeteoSwiss	1983-2017
40	Frasco (CH)	705180	132790	890	UCA	1983-2007
41	Fusio (CH)	694115	144405	1276	UCA	1983-2017
42	Fusio Diga Sambuco (CH)	693682	145597	1471	UCA	1998-2007
43	Giubiasco (CH)	719712	114774	215	UCA	1988-2017

N°	Sampling site	Longitude (m)	Latitude (m)	Altitude (m a.s.l.)	Date source	Period
44	Gnosca (CH)	721880	122072	247	UCA	1983-2017
45	Göschenen (CH)	688477	171926	950	MeteoSwiss	1988-2017
46	Göscheneralp (CH)	681250	166790	1745	MeteoSwiss	1988-2017
47	Grancia (CH) (CH)	715328	92408	310	UCA	2008-2017
48	Grimsel Hospiz (CH)	668583	158215	1980	MeteoSwiss	1983-2017
49	Grono (CH)	733017	124090	324	MeteoSwiss	1983-2017
50	Gütsch ob Andermatt (CH)	690050	167475	2287	MeteoSwiss	1983-2017
51	Hinterrhein (CH)	733900	153980	1611	MeteoSwiss	1988-2012
52	Isone (CH)	720176	110336	792	UCA	1983-2017
53	Lago Delio (I)	701480	103619	835	ARPA Piemonte	1983-1992
54	Lago Morasco (I)	673975	142207	1820	ENEL	1983-2017
55	Lago Sabbione (I)	670084	141680	2462	ENEL	1983-2017
56	Lago Toggia (I)	676351	143398	2200	ENEL	1983-2017
57	Lago Truzzo (I)	744807	136010	2064	ARPA Lombardia	1983-1987, 2008-2012
58	Lavena Ponte Tresa (I)	710314	91714	279	ARPA Lombardia	1983-2017
59	Locarno Monti (CH)	704160	114350	367	MeteoSwiss	1983-2017
60	Lodrino (CH)	719843	128306	258	UCA	1983-2012
61	Lugano (CH)	717874	95884	273	MeteoSwiss	1983-2017
62	Luino (I)	701193	94821	205	ARPA Lombardia	2008-2017
63	Magadino/Aeroporto (CH)	711170	113542	197	MeteoSwiss	1983-2007
64	Magadino/Cadenazzo (CH)	715475	113162	203	MeteoSwiss	2008-2017
65	Maggia (CH)	697620	122190	316	UCA	1983-2017
66	Mendrisio (CH)	719211	82996	290	UCA	1983-2012
67	Mesocco (CH)	737850	139825	830	MeteoSwiss	1983-2017
68	Moleno (CH)	719940	126559	255	OASI	2008-2017
69	Monte Brè (CH)	720679	96765	910		1983-1987
70	Monte Generoso (CH)	722503	87456	1600	MeteoSwiss	2003-2017
71	Morbio Superiore (CH)	722750	80075	440	MeteoSwiss	1983-2017
72	Mosogno (CH)	692803	117050	771	MeteoSwiss	1983-2017
73	Novaggio (CH)	709980	96160	620	UCA	2008-2017
74	Oberwald (CH)	669650	154050	1375	MeteoSwiss	1983-2002
75	Olivone (CH)	715465	154865	958	MeteoSwiss	1983-2017
76	Olivone (CH)	715410	154120	909	UCA	1983-2017
77	Olivone Luzzone Diga (CH)	716665	158232	1612	UCA	1998-2017
78	Piano dei Camosci (I)	670990	143013	2450	ARPA Piemonte	1998-2017
79	Piotta (CH)	695888	152261	990	MeteoSwiss	1983-2017
80	Ponte Formazza (I)	676012	136332	1300	ENEL	1983-2012
81	Ponte Tresa (CH)	710110	91630	274	MeteoSwiss	2008-2017
82	Robiei (CH)	682588	144091	1896	MeteoSwiss	1983-2017
83	S. Antonino (CH)	718440	113030	215	UCA	1983-1987
84	S. Bernardino (CH)	734112	147296	1639	MeteoSwiss	1983-2017
85	Scudellate (CH)	724175	86850	925	MeteoSwiss	1983-2017
86	Sedrun (CH)	699974	169845	1429	MeteoSwiss	1983-2017
87	Somazzo (CH)	720490	81758	527	UCA	1998-2012
88	Sonogno (CH)	703822	134055	912	MeteoSwiss	1983-2017
89	Sonogno (CH)	703633	133902	913	UCA	2013-2017
90	Splügen (CH)	744420	157435	1460	MeteoSwiss	1983-2017
91	Stabio (CH)	716050	77966	353	MeteoSwiss	1983-2017
92	Ulrichen (CH)	666740	150760	1346	MeteoSwiss	1983-2007
93	Vals (CH)	734016	165552	1278	MeteoSwiss	1983-2017
94	Vira Gambarogno (CH)	709400	111680	199	MeteoSwiss	1983-2017
95	Vrin (CH)	727220	168526	1384	MeteoSwiss	1983-2017
96	Zervreila (CH)	728780	160000	1738	MeteoSwiss	1983-2017

Table A2: Mean annual concentrations in wet deposition sampling sites. Prec and Cond correspond to precipitation and conductivity, respectively.

Year	Prec	Analysed Prec	Cond 20°C	pH	Ca^{2+}	Mg^{2+}	Na^+	K^+	NH_4^+	NO_3^-	SO_4^{2+}	Cl^-	Acidity
	mm	%	µS cm^{-1}						meq m^{-3}				
Acquarossa													
1990	1056	44	25	4.4	16	6	19	5	36	41	52	30	41
1991	1201	42	15	4.7	7	4	15	2	29	29	33	8	19
1993	1746	79	15	4.8	32	6	8	2	30	29	39	12	8
1994	1432	91	14	5.0	31	12	7	1	33	29	42	8	-2
1995	1044	56	13	4.8	19	8	4	3	28	30	35	4	13
1996	1282	72	13	4.9	26	6	6	1	31	28	30	8	-3
1997	1083	77	10	5.0	11	3	4	1	22	23	24	5	10
1998	1227	80	13	4.9	17	5	6	1	34	30	31	5	9
1999	1688	72	12	5.2	32	9	5	2	35	29	37	5	-5
2000	1684	85	14	5.4	62	12	8	2	23	25	40	7	-31
2001	1344	88	12	5.1	23	9	5	1	37	33	38	4	3
2002	1792	87	12	5.2	42	7	10	7	23	20	34	4	-28
2003	923	77	12	5.5	34	8	6	2	36	31	32	5	-14
2004	1241	64	9	5.5	23	6	7	3	34	24	22	5	-12
2005	661	74	16	5.8	48	14	6	4	58	45	40	5	-28
2006	1018	31	7	5.3	17	6	4	1	12	18	15	4	4
2007	954	85	11	5.4	24	7	6	2	38	30	33	4	-7
2008	1696	55	7	5.3	17	5	4	1	18	22	17	4	2
2009	1355	91	7	5.6	18	6	3	1	23	22	18	4	-5
2010	1388	89	8	5.5	23	7	3	1	28	22	19	3	-12
2011	1048	88	8	5.6	19	5	5	2	29	23	19	5	-11
2012	1507	97	10	5.5	20	3	8	2	35	27	19	7	-20
2013	1393	76	10	5.4	23	4	5	2	38	27	19	5	-19
2014	1678	95	6	5.3	17	5	3	1	15	15	8	3	-12
2015	1157	93	9	5.3	17	5	5	2	34	26	16	6	-13
2016	1124	91	9	5.4	23	4	6	2	36	29	15	5	-21
2017	1094	90	7	5.7	10	2	4	2	36	23	12	4	-15
Bignasco													
2002	2564	69	10	5.0	13	3	8	4	21	22	25	6	7
2003	1087	92	12	5.1	21	3	6	3	39	32	30	5	-3
2004	1663	14	12	4.8	18	2	5	3	24	36	21	5	12
2005	1030	77	13	5.1	20	3	5	2	47	37	27	4	0
2006	1308	52	14	5.0	19	4	10	2	38	39	25	6	6
2007	1148	101	11	5.1	16	4	7	3	28	29	26	6	5
2008	1983	101	9	5.1	20	3	7	2	23	24	18	8	-1
2009	1945	89	8	5.2	11	2	7	3	21	21	14	6	2
2010	1544	93	7	5.2	12	2	5	2	20	21	15	4	3
2011	1302	97	8	5.4	15	3	4	1	26	25	17	4	-1
2012	1684	98	9	5.4	15	4	7	2	31	27	16	7	-9
2013	1737	106	8	5.4	15	2	5	2	26	22	13	5	-11
2014	2107	93	7	5.3	18	3	4	2	18	20	11	4	-10
2015	1287	104	9	5.3	19	3	8	2	29	28	16	7	-9
2016	1470	103	9	5.4	17	3	5	2	30	25	13	5	-15
2017	1459	89	8	5.5	10	2	6	2	31	25	12	6	-8

Year	Prec mm	Analysed Prec %	Cond 20°C µS cm^{-1}	pH	Ca^{2+}	Mg^{2+}	Na$^+$	K$^+$	NH$_4^+$	NO$_3^-$	SO$_4^{2+}$	Cl$^-$	Acidity
								meq m^{-3}					
Monte Brè													
1996	1721	75	14	4.7	14	4	5	1	34	31	35	5	17
1997	1227	76	14	4.9	14	4	5	1	36	33	35	6	13
1998	1508	84	15	4.9	23	5	9	2	43	36	40	6	9
1999	1699	90	15	5.1	38	7	10	3	46	38	44	10	0
2000	2149	90	12	5.2	29	6	10	2	29	28	31	7	-6
2001	1499	87	17	4.9	21	5	11	2	54	44	46	9	10
2002	2105	98	16	5.1	64	6	8	3	33	30	33	9	-40
2003	1167	88	15	5.2	25	5	10	4	54	45	32	9	-9
2004	1373	19	15	4.9	20	4	12	3	40	45	28	8	13
2005	947	92	24	4.8	29	6	20	5	75	69	41	20	7
2006	1304	89	15	5.2	31	5	11	2	51	43	33	7	-8
2007	1143	94	11	5.3	15	3	8	2	41	33	26	7	-4
2008	2188	89	11	5.2	17	5	9	2	32	31	25	9	1
2009	1680	79	10	5.3	21	5	6	3	31	33	19	7	-5
2010	1854	79	12	5.3	40	9	9	6	26	33	33	8	-10
2011	1179	52	9	5.2	17	4	6	2	23	24	16	6	-4
2012	1412	96	10	5.4	13	4	8	2	35	29	17	8	-9
2013	1713	89	10	5.3	16	4	7	5	26	26	13	8	-12
2014	2430	93	7	5.5	16	3	5	2	23	20	12	5	-12
2015	1232	92	11	5.6	19	5	10	3	42	34	20	10	-18
2016	1681	86	9	5.6	20	3	7	3	34	26	14	9	-19
2017	1506	93	10	5.8	13	3	9	4	44	30	16	11	-17
Locarno Monti													
1988	2127	66	25	4.4	29	5	10	2	45	38	73	11	37
1989	1478	73	25	4.3	20	5	13	4	31	37	62	15	45
1990	1474	62	34	4.3	19	6	21	6	58	61	72	35	53
1991	1782	50	19	4.6	9	4	21	3	38	33	44	14	25
1993	2134	80	19	4.6	29	7	11	2	37	34	50	13	20
1994	1865	81	18	4.7	25	9	8	1	34	30	51	9	14
1995	1406	86	19	4.6	28	9	6	2	38	35	58	9	21
1996	1661	72	20	4.6	26	8	9	1	41	39	48	10	13
1997	1564	90	15	4.8	16	5	7	2	34	32	38	9	16
1998	1752	83	19	4.7	23	5	10	3	52	45	47	8	15
1999	2367	76	17	4.9	33	6	9	2	50	43	46	8	5
2000	2623	83	17	4.9	49	8	11	2	40	38	42	9	-15
2001	1711	86	20	4.7	23	5	9	1	56	49	49	8	15
2002	2646	94	15	5.0	44	4	6	2	38	30	30	6	-26
2003	1107	83	15	4.9	21	3	7	2	48	42	35	6	4
2004	1894	11	8	5.0	11	2	5	1	14	17	16	2	9
2005	1057	88	17	5.0	28	4	8	2	62	50	36	5	-2
2006	1629	72	14	5.0	19	4	11	2	40	36	29	9	5
2007	1501	87	12	5.1	15	3	5	1	45	37	29	5	1
2008	2346	92	11	5.1	17	3	7	1	32	32	24	7	3
2009	2092	94	10	5.2	15	3	5	1	36	29	22	5	-3
2010	2020	86	10	5.1	17	3	5	1	32	30	21	5	1
2011	1578	91	11	5.3	21	4	6	2	39	34	26	5	-3
2012	1863	89	11	5.3	15	4	7	2	37	32	20	7	-6
2013	1947	88	9	5.3	15	3	5	1	30	25	15	5	-9
2014	2781	85	8	5.3	17	3	5	2	23	21	13	4	-10
2015	1345	89	12	5.2	17	3	8	1	45	36	21	8	-10
2016	1620	91	11	5.5	23	4	9	2	43	33	19	8	-22
2017	1672	93	9	5.6	11	3	7	1	40	27	15	7	-15

Year	Prec	Analysed Prec	Cond 20°C	pH	Ca^{2+}	Mg^{2+}	Na$^+$	K$^+$	NH$_4^+$	NO$_3^-$	SO$_4^{2+}$	Cl$^-$	Acidity
	mm	%	µS cm^{-1}		meq m^{-3}								
Lugano													
1989	1468	68	32	4.3	27	8	19	5	48	55	84	19	54
1990	1342	57	32	4.4	37	8	19	6	74	63	81	32	34
1991	1288	39	26	4.5	18	7	26	7	52	56	62	14	35
1993	1590	75	20	4.6	30	7	11	3	45	41	59	13	22
1994	1703	82	18	4.8	31	12	11	2	43	37	58	12	8
1995	1412	77	18	4.7	26	10	8	2	40	36	55	8	16
1996	1721	64	16	4.8	27	8	10	1	39	33	45	11	6
1997	1227	97	16	4.8	19	6	8	2	44	38	43	9	13
1998	1508	78	19	4.8	27	6	10	3	51	43	49	9	9
1999	1699	66	20	4.8	31	7	10	2	59	48	54	11	5
2000	2149	86	16	5.0	38	7	11	2	41	40	42	9	-5
2001	1498	66	18	4.8	22	6	13	4	55	47	51	11	13
2002	2099	91	19	5.0	70	7	12	5	45	38	49	11	-36
2003	1181	77	23	5.1	49	9	14	5	73	67	56	12	-18
2004	1373	49	11	5.5	25	5	7	3	42	32	26	6	-10
2005	947	54	19	5.1	36	7	10	5	67	61	42	9	-6
2006	1304	56	19	5.2	42	12	17	4	67	54	46	16	-13
2007	1143	66	14	5.5	29	7	13	6	43	39	35	9	-15
2008	2188	77	12	5.3	24	6	10	2	41	36	33	10	-4
2009	1680	73	11	5.4	25	5	6	2	39	33	23	7	-11
2010	1854	81	18	5.4	39	8	30	34	33	34	92	9	-6
2011	1179	77	11	5.4	19	4	5	4	42	34	23	5	-10
2012	1412	81	11	5.4	14	4	9	2	41	32	20	9	-10
2013	1713	74	10	5.4	15	3	7	2	35	28	16	7	-13
2014	2430	73	7	5.5	14	3	5	2	23	19	13	5	-8
2015	1232	77	11	5.4	14	3	8	2	45	32	19	8	-15
2016	1681	78	9	5.6	13	2	5	2	38	27	15	6	-15
2017	1506	81	9	5.6	11	3	7	2	45	30	17	7	-14
Piotta													
1990	1281	41	22	4.4	15	4	17	4	28	35	46	25	38
1991	1475	30	22	4.6	40	6	22	5	36	38	45	23	8
1993	2005	84	12	4.8	15	3	6	1	22	22	31	9	16
1994	1279	80	13	4.9	30	9	9	1	26	26	34	10	0
1995	1235	78	11	5.0	20	6	9	1	21	23	32	10	9
1996	1597	69	10	5.0	13	5	8	1	26	22	24	8	7
1997	1345	73	10	5.0	11	3	9	1	18	22	21	9	11
1998	1426	69	12	4.9	13	3	8	1	30	26	29	6	11
1999	1722	78	11	5.1	21	4	5	1	29	25	28	5	3
2000	1853	83	14	5.1	63	6	9	1	23	24	29	8	-43
2001	1505	89	12	4.9	14	3	9	1	33	28	28	8	7
2002	2017	94	7	5.2	14	2	4	1	20	16	15	3	-3
2003	1061	87	12	5.3	20	4	9	2	44	29	26	9	-16
2004	1118	73	8	5.4	14	3	13	2	21	17	13	11	-8
2005	644	101	11	5.3	14	2	6	1	39	30	21	6	-4
2006	1176	78	9	5.2	16	3	9	1	24	24	17	8	2
2007	1313	80	9	5.3	12	2	5	1	29	23	20	5	-1
2008	1803	85	9	5.3	16	4	11	1	25	23	18	10	-4
2009	1360	89	7	5.4	11	2	7	1	22	19	14	7	-1
2010	1377	83	9	5.3	13	3	9	3	26	20	14	9	-7
2011	1269	91	7	5.5	14	3	9	2	23	18	15	8	-5
2012	1602	85	8	5.6	15	3	8	2	32	20	15	7	-16
2013	1692	86	7	5.5	14	2	7	1	21	16	12	7	-10
2014	1813	84	6	5.4	16	3	6	2	15	12	8	6	-14
2015	1189	78	8	5.6	14	3	7	4	31	19	16	7	-15
2016	1234	89	7	5.5	16	2	6	1	25	20	11	6	-13
2017	1287	69	8	5.5	10	2	12	2	33	22	12	13	-11

Year	Prec	Analysed Prec	Cond 20°C	pH	Ca^{2+}	Mg^{2+}	Na$^+$	K$^+$	NH$_4^+$	NO$_3^-$	SO$_4^{2+}$	Cl$^-$	Acidity
	mm	%	µS cm^{-1}		meq m^{-3}								
Robiei													
1997	2202	88	8	5.0	10	2	3	1	15	15	15	3	10
1998	2216	90	10	5.0	15	3	6	1	21	19	19	3	6
1999	2845	68	10	5.2	22	7	6	1	30	25	25	4	-2
2000	3330	86	11	5.2	28	5	6	1	25	24	24	4	-6
2001	2258	92	12	4.9	13	3	5	1	32	28	28	3	11
2002	2782	72	9	5.1	10	2	3	1	27	21	21	2	2
2003	1625	91	9	5.3	12	2	3	1	37	29	29	2	-2
2004	2277	42	6	5.3	11	2	4	1	20	17	17	3	-2
2005	1357	78	12	5.1	17	2	2	1	43	34	34	2	-1
2006	2145	77	8	5.2	13	2	3	1	25	24	24	3	4
2007	1639	96	9	5.2	11	2	3	1	29	24	24	3	1
2008	3188	62	9	5.2	21	3	4	1	26	24	24	4	-5
2009	2094	46	6	5.5	15	2	3	1	16	18	18	5	0
2010	2599	75	6	5.3	11	6	2	1	17	18	18	2	-1
2011	2071	80	7	5.5	21	2	2	1	21	19	19	2	-10
2012	2721	77	8	5.5	10	2	3	1	30	23	23	3	-6
2013	2601	80	7	5.4	13	2	3	1	19	18	18	3	-6
2014	2802	72	7	5.5	14	2	3	1	17	17	17	2	-8
2015	2303	82	9	5.4	14	3	4	1	27	29	29	4	-2
2016	2341	76	8	5.6	15	2	4	1	25	23	23	3	-11
2017	2059	79	9	5.1	11	2	3	1	22	28	28	3	5
Sonogno													
2002	2782	87	11	5.1	17	5	6	6	26	22	22	4	-11
2003	1435	76	11	5.2	20	3	6	2	35	31	26	6	-2
2004	1980	37	9	5.4	12	3	8	2	36	26	19	5	-4
2005	1033	42	11	5.2	21	2	5	1	29	26	22	4	-2
2006	1858	66	12	5.1	22	9	11	4	26	29	24	5	-6
2007	1659	84	10	5.3	15	3	6	3	40	30	28	4	-4
2008	2957	80	8	5.5	16	3	7	2	22	21	16	7	-5
2009	2033	95	8	5.5	14	3	6	2	25	23	15	6	-4
2010	2116	90	9	5.5	19	6	5	2	30	22	17	4	-16
2011	1969	75	8	5.5	13	3	5	2	29	22	16	4	-8
2012	2317	86	10	5.6	15	3	8	3	35	28	17	8	-13
2013	2003	83	8	5.8	16	2	7	4	32	21	13	5	-24
2014	2604	93	8	5.7	22	6	7	3	23	17	12	5	-25
2015	1694	85	10	5.6	17	3	9	3	44	32	18	8	-17
2016	1849	95	9	5.8	19	3	6	2	36	25	13	7	-26
2017	1772	92	7	5.8	9	2	5	2	30	21	10	5	-14

Year	Prec	Analysed Prec	Cond 20°C	pH	Ca²⁺	Mg²⁺	Na⁺	K⁺	NH₄⁺	NO₃⁻	SO₄²⁺	Cl⁻	Acidity
	mm	%	µS cm⁻¹						meq m⁻³				
Stabio													
1991	1181	56	32	4.5	38	10	30	9	75	69	88	23	34
1993	1394	64	22	4.8	47	13	13	4	55	49	74	15	12
1994	1485	91	21	4.8	39	12	13	2	55	45	66	14	6
1995	1487	81	22	4.6	26	8	10	1	54	46	61	10	20
1996	1706	61	18	4.8	29	8	12	2	45	38	47	10	6
1997	1269	87	16	4.9	20	6	8	1	50	37	44	9	9
1998	1566	90	17	4.9	25	7	10	2	57	41	47	11	6
1999	1410	89	19	4.9	35	8	12	2	59	48	52	11	1
2000	2336	88	16	5.0	33	7	11	2	49	40	43	9	-7
2001	1251	97	19	4.9	29	6	11	2	63	52	52	10	4
2002	2438	77	19	5.1	67	6	10	2	54	36	42	11	-46
2003	1206	90	15	5.0	19	4	8	2	54	43	32	7	-5
2004	1364	69	12	5.3	21	4	8	4	48	38	31	6	-7
2005	898	95	17	5.2	26	5	9	3	75	55	36	9	-11
2006	1184	90	15	5.2	24	5	13	2	62	48	33	10	-6
2007	1183	88	13	5.4	15	4	10	4	52	38	31	8	-8
2008	2121	93	11	5.3	15	4	9	3	40	32	22	9	-6
2009	1613	99	10	5.3	14	4	8	2	36	30	19	8	-5
2010	2132	92	10	5.3	16	3	7	2	40	32	21	6	-8
2011	1239	92	12	5.4	20	4	7	2	49	36	24	6	-12
2012	1443	96	10	5.5	14	4	9	2	37	29	17	8	-12
2013	1518	100	11	5.4	13	3	8	2	40	29	17	7	-14
2014	2549	92	8	5.5	15	3	6	1	29	22	13	5	-13
2015	1414	97	12	5.4	16	4	8	2	49	33	22	7	-18
2016	1763	89	11	5.7	16	3	7	2	50	32	18	7	-24
2017	1372	92	10	5.6	11	2	7	2	49	33	16	7	-20

Table A3: Average concentrations in rainwater in the periods of 1983-1987, 1988-1992, 1993-1997, 1998-2002, 2003-2007, 2008-2012, 2013-2017 used for the correlation analysis. Prec corresponds to precipitation. Values in red are based completely on estimated yearly mean concentrations, values in green only partially.

Period	Prec	SO_4^{2+}	NO_3^-	$NH4^+$	BC
	mm	meq m^{-3}			
Acuarossa					
1983-1992	1333	52	30	33	45
1988-1992	1143	45	31	31	30
1993-1997	1317	35	28	29	34
1998-2002	1547	36	27	30	48
2003-2007	959	29	30	38	38
2008-2012	1399	18	23	26	27
2013-2017	1289	14	24	30	23
Bignasco					
1983-1987	1808	59	31	31	29
1988-1992	1552	50	32	30	24
1993-1997	1917	41	20	27	27
1998-2002	2144	34	31	32	28
2003-2007	1247	27	33	36	25
2008-2012	1691	16	24	24	20
2013-2017	1612	13	23	26	20
Monte Brè					
1983-1987	1585	59	38	41	36
1988-1992	1451	53	41	44	32
1993-1997	1530	40	33	35	28
1998-2002	1791	38	34	40	45
2003-2007	1189	30	42	50	32
2008-2012	1663	20	30	31	24
2013-2017	1712	14	26	32	23
Locarno Monti					
1983-1987	1968	71	40	42	39
1988-1992	1701	61	41	42	29
1993-1997	1726	49	34	37	34
1998-2002	2220	42	40	46	44
2003-2007	1438	32	40	46	26
2008-2012	1980	22	31	35	22
2013-2017	1873	16	27	34	21
Lugano					
1983-1987	273	80	44	49	45
1988-1992	1451	72	48	53	39
1993-1997	1530	52	37	42	38
1998-2002	1791	48	43	49	50
2003-2007	1189	41	50	58	48
2008-2012	1663	25	34	40	29
2013-2017	1712	15	26	35	18
Piotta					
1983-1987	1400	41	24	28	29
1988-1992	1375	35	25	28	22
1993-1997	1492	29	23	23	23
1998-2002	1705	25	23	26	31
2003-2007	1062	19	24	30	19
2008-2012	1482	15	20	26	18
2013-2017	1443	11	17	24	19

Period	Prec	SO$_4$$^{2+}$	NO$_3$$^-$	NH4$^+$	BC
	mm	meq m^{-3}			
Robiei					
1983-1987	2688	49	29	39	31
1988-1992	2362	42	30	39	23
1993-1997	2476	34	24	31	26
1998-2002	2686	27	24	27	24
2003-2007	1808	22	28	34	18
2008-2012	2535	15	22	25	19
2013-2017	2403	12	22	22	17
Sonogno					
1983-1987	2410	52	28	33	30
1988-1992	1821	45	29	33	26
1993-1997	2245	36	26	31	28
1998-2002	2446	31	28	33	30
2003-2007	1593	25	30	34	27
2008-2012	2278	16	23	28	22
2013-2017	2028	13	22	32	24
Stabio					
1983-1987	1430	83	43	52	53
1988-1992	1381	73	50	57	41
1993-1997	1468	58	43	51	43
1998-2002	1800	46	42	55	49
2003-2007	1167	32	44	57	28
2008-2012	1709	21	32	40	21
2013-2017	1723	17	29	42	19
Bellinzago (Italy)					
1983-1987	1059	89	49	64	58
1988-1992	951	84	60	78	44
1993-1997	1135	57	46	53	31
1998-2002	1155	50	46	62	71
2003-2007	849	36	43	66	30
2008-2012	1198	27	35	56	31
2013-2017	1171	24	37	56	39
Devero (Italy)					
1983-1987	1762	46	24	27	24
1988-1992	1592	39	25	25	22
1993-1997	1779	30	21	23	19
1998-2002	2070	24	23	26	24
2003-2007	1180	20	25	31	20
2008-2012	1771	13	16	21	14
2013-2017	1661	11	15	21	15
Domodossola (Italy)					
1983-1987	1403	55	30	34	28
1988-1992	1273	46	32	31	26
1993-1997	1475	35	27	28	22
1998-2002	1622	27	27	31	29
2003-2007	1045	23	33	42	19
2008-2012	1489	14	22	26	18
2013-2017	1336	13	20	28	18
Graniga (Italy)					
1983-1987	1429	56	31	35	26
1988-1992	1621	47	32	33	24
1993-1997	1806	37	28	31	23
1998-2002	1950	28	29	32	26
2003-2007	1309	23	32	42	19
2008-2012	1689	15	23	27	17
2013-2017	1637	14	23	30	18

Period	Prec	SO$_4^{2+}$	NO$_3^-$	NH4$^+$	BC
	mm	meq m^{-3}			
Lunecco (Italy)					
1983-1987	2204	62	37	43	34
1988-1992	2046	54	38	46	27
1993-1997	2304	44	34	39	25
1998-2002	2670	37	35	43	32
2003-2007	1657	27	36	45	18
2008-2012	2352	18	27	34	19
2013-2017	2270	16	24	35	29
Pallanza (Italy)					
1983-1987	1591	81	46	57	39
1988-1992	1735	69	49	61	30
1993-1997	1972	57	44	52	28
1998-2002	2189	44	43	56	34
2003-2007	1426	34	46	65	21
2008-2012	1982	21	32	44	16
2013-2017	1950	17	29	43	20
S. Monte Orta (Italy)					
1983-1987	1468	84	49	58	43
1988-1992	1412	74	55	66	35
1993-1997	1725	55	44	53	29
1998-2002	1900	44	44	55	37
Monte Mesma (Italy)					
2003-2007	1257	34	47	59	24
2008-2012	1760	22	35	48	21
2013-2017	1682	19	33	50	26
Basodino (Glacier)					
1983-1987	2688	20	12	14	18
1988-1992	2362	18	11	14	14
1993-1997	2476	20	10	14	30
1998-2002	2686	10	8	7	15
2003-2007	1808	9	11	13	13
2008-2012	2535	9	13	14	9
2013-2017	2403	6	10	10	13

Table A4: Results from multiple regression analysis for different time periods. n, r^2, F, p stay for data number, coefficient of determination, F statistic and p-values.

Period	n	r^2	F	p	m_{lat} meq m^{-4}	m_{long} meq m^{-4}	m_{alt} meq m^{-4}	C_0 meq m^{-3}
Base cations								
1983-1987	17	0.74	16.4	0.000	2.5E-4	-1.9E-4	-3.0E-3	-115.7
1988-1992	17	0.86	35.0	0.000	-1.7E-4	1.5E-4	-2.4E-3	-56.8
1993-1997	17	0.55	7.6	0.003	-7.2E-5	2.4E-4	1.4E-3	-130.6
1998-2002	17	0.81	23.3	0.000	-1.5E-4	3.5E-4	-4.2E-3	184.5
2003-2007	17	0.53	7.0	0.005	-7.8E-6	2.9E-4	-3.1E-3	-171.6
2008-2012	17	0.59	8.6	0.002	-2.9E-5	1.4E-4	-2.8E-3	-71.6
2013-2017	17	0.34	3.8	0.038	-1.0E-4	4.4E-5	-1.7E-3	3.5
Ammonium								
1983-1987	17	0.80	21.9	0.000	-3.1E-4	6.2E-5	-4.7E-3	35.6
1988-1992	17	0.81	23.7	0.000	-4.5E-4	1.3E-4	-3.1E-3	3.2
1993-1997	17	0.80	21.9	0.000	-2.9E-4	8.8E-5	-3.6E-3	9.2
1998-2002	17	0.89	43.0	0.000	-3.0E-4	1.0E-4	-7.6E-3	6.0
2003-2007	17	0.89	42.1	0.000	-3.0E-4	7.2E-5	-7.4E-3	33.6
2008-2012	17	0.81	24.1	0.000	-2.7E-4	7.4E-5	-3.2E-3	13.2
2013-2017	17	0.85	30.2	0.000	-2.4E-4	4.7E-5	-5.7E-3	31.1
Sulphate								
1983-1987	17	0.87	35.6	0.000	-3.6E-4	9.5E-5	-1.1E-2	44.4
1988-1992	17	0.88	41.3	0.000	-3.7E-4	1.3E-4	-8.2E-3	9.4
1993-1997	17	0.83	27.5	0.000	-2.4E-4	9.7E-5	-5.5E-3	5.0
1998-2002	17	0.88	40.6	0.000	-1.6E-4	1.8E-4	-6.8E-3	-64.5
2003-2007	17	0.82	24.8	0.000	-1.0E-4	1.2E-4	-5.1E-3	-41.5
2008-2012	17	0.80	23.0	0.000	-7.5E-5	9.4E-5	-2.1E-3	-36.8
2013-2017	17	0.81	23.1	0.000	-8.2E-5	1.4E-5	-1.9E-3	15.8
Nitrate								
1983-1987	17	0.86	34.0	0.000	-2.0E-4	7.2E-5	-5.5E-3	11.5
1988-1992	17	0.90	47.1	0.000	-2.9E-4	9.5E-5	-5.2E-3	7.8
1993-1997	17	0.88	39.0	0.000	-2.0E-4	6.0E-5	-5.3E-3	16.2
1998-2002	17	0.88	39.3	0.000	-1.8E-4	7.7E-5	-6.7E-3	4.2
2003-2007	17	0.83	27.0	0.000	-1.7E-4	1.0E-4	-6.5E-3	-10.2
2008-2012	17	0.85	31.2	0.000	-1.4E-4	1.0E-4	-2.8E-3	-27.9
2013-2017	17	0.78	19.4	0.000	-1.3E-4	5.4E-5	-3.1E-3	3.1