3. Acidifying deposition in Southern Switzerland. Assessment of the trend 1988-2007

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The sulphur dioxide and nitrogen oxides emissions in Switzerland started to increase steeply after the second world war. Sulphur dioxide reached its maximum between 1965 and 1980, while nitrogen oxides peaked around 1985. Afterwards, both sulphur and nitrogen oxides decreased continuously until 2000 (see figure A). For ammonia only a small decrease could be observed. Because of its particular meteorology the air quality in Southern Switzerland is not only influenced by the local emissions but also by transboundary air pollution originating from the Po Plain and particularly from the heavily polluted urban area of Milan. Furthermore, many high altitude soils and freshwaters of Southern Switzerland are particularly sensitive to acidification due to the dominance of base-poor rocks with low buffering capacity. As a consequence, acidifying deposition in Southern Switzerland becomes particularly relevant.

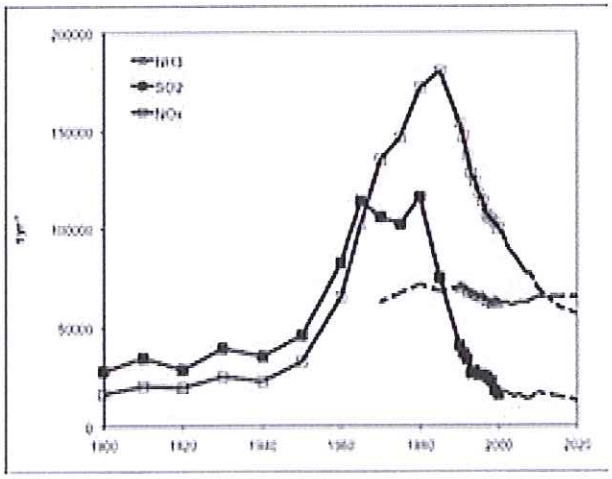


Figure A *Annual sulphur dioxide, nitrogen oxides and ammonia emissions in Switzerland from 1900 to 2010. Dashed lines indicate estimate values.*

It is for this reason that rainwater quality has been studied in Southern Switzerland since the beginning of the eighties, first only at Locarno Monti and Lugano and later also at other sampling stations. Nowadays weekly precipitation is collected at 9 sampling sites distributed over the whole area of Canton Ticino. A trend analysis of the main chemical parameters in rainwater reveals that sulphate concentrations have decreased, reflecting the decrease of SO_2 emissions after 1980. At Locarno Monti and Lugano sulphate concentrations have decreased by around 57-66% during the last 20 years, while no significant trend could be observed for ammonium and nitrate concentrations. Concentrations of bicarbonate and base cations have increased and were particularly high during the years 1999, 2000, 2002, when precipitation was higher than usual and alkaline rain events were more frequent. As a consequence of decreased sulphate concentrations and increased concentrations of base cations, acidity of rainwater decreased and therefore pH values increased. Since the end of the eighties until the beginning of this millennium the mean annual pH values of rainwater at Locarno Monti and

Lugano increased from 4.3 to 5.0/5.3 and after 2000 the mean annual acidity became negative at most sites.

In order to analyze patterns in the data and relations between parameters, a principal component analysis has been performed with average concentrations of the main chemical parameters and with conductivity, precipitation amounts and geographic parameters over the periods of 1988-1992, 1993-1997, 1998-2002 and 2003-2007. The analysis has been extended by considering not only data from Swiss monitoring stations but also from North Italian sampling sites. The analysis has shown that most chemical parameters in rainwater correlate significantly with the geographic parameters latitude, longitude and altitude. Sulphate, nitrate and ammonium concentrations correlate with both latitude and altitude reflecting the transport of these pollutants from the urban area of Milan and from rural areas of the Po Plain towards north. Conversely, mean yearly concentrations of base cations and bicarbonate, which increase with decreasing annual precipitation (smaller dilution of alkaline rain events on an annual base), correlate with longitude because atmospheric currents causing rainfall are mostly directed from south-west to north-east. The principal component analysis also reveals that acidity correlated with latitude during the beginning of the monitored period (1988-1992), but became more and more independent of latitude and dependent of longitude after the interval of 1998-2002. This indicates that acidity was mainly determined by emissions of SO_2, NO_x and NH_3 during the first period, while alkaline rain events became more important afterwards.

The geographic distribution of chemical parameters in rainwater discussed in the principal component analysis suggested the possibility to develop a multiple regression model with the variables latitude, longitude, altitude. This can describe the geographic distribution of the concentrations of single chemical parameters in rainwater. Multiple linear regression analyses have been performed for different parameters in the periods of 1988-1992, 1993-1997, 1998-2002 and 2003-2007. For some parameters and time periods the addition of the mean annual precipitation volume as a regression variable has provided better results. In particular, parameters like base cations, bicarbonate, acidity and pH, whose annual mean concentrations are strongly influenced by sporadic alkaline rain events, are better modeled after 1993 if the amount of precipitation is also considered. The regression parameters have then been used to map concentrations of the main chemical parameters over Southern Switzerland with a resolution of 1km x 1km.

Finally, wet deposition maps for Southern Switzerland have been derived by multiplying the concentration maps of chemical parameters in rainwater by the precipitation maps (see figure B). Observations made for the concentrations of chemical parameters in rainwater do not substantially differ from those of wet deposition. In general time trends can be observed, in which sulphate deposition and acidity decrease, whereas the deposition of base cations and of bicarbonate increase. The dependency of the chemical variables on geographic parameters also did not vary over time. However, the particularly rain rich (1998-2002) and rain poor (2003-2007) years had visible consequences on deposition: wet deposition of sulphate, nitrate and ammonium were slightly higher between 1998 and 2002 compared to the immediately previous and subsequent time periods.

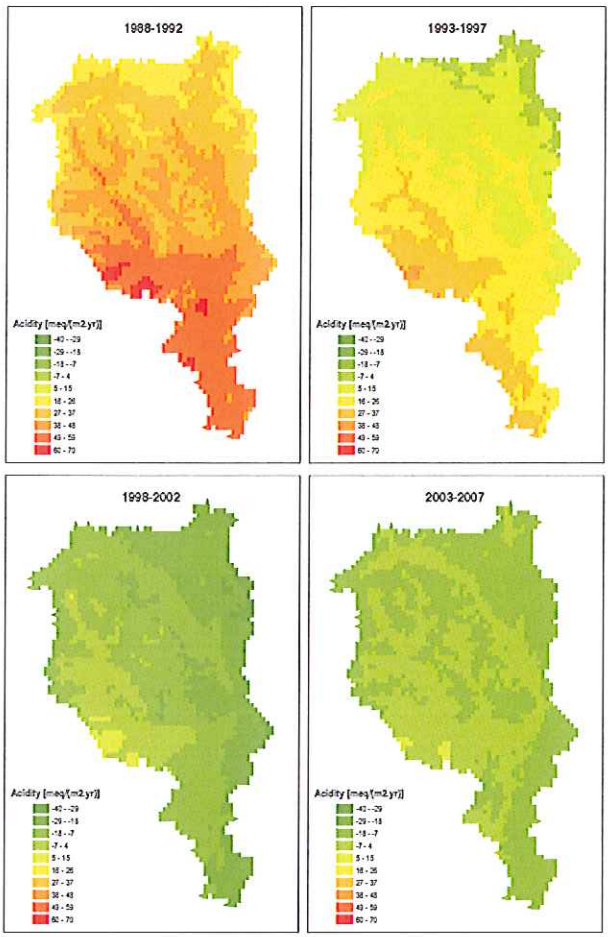
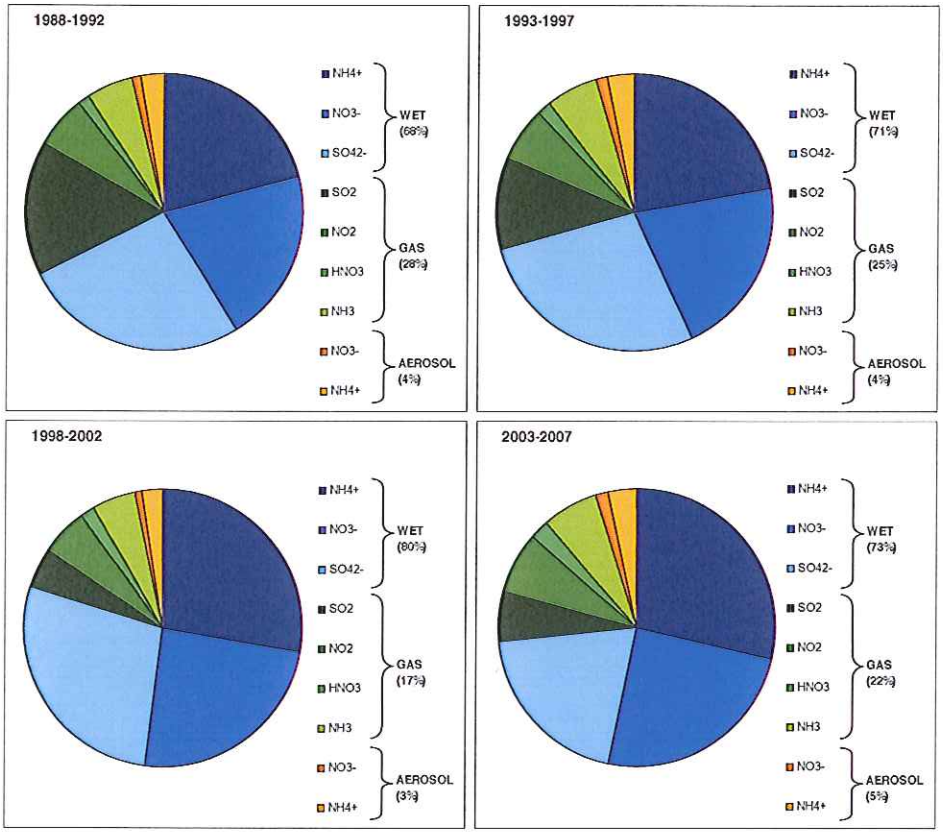


Figure B. *Wet deposition: 1988-1992, 1993-1997, 1998-2002, 2003-2007. Deposition of acidity*

In addition to wet deposition the dry deposition of gases and aerosols also contribute to the total acidifying deposition. Maps of dry deposition were prepared by Meteotest. The sum of wet and dry deposition was used to derive maps of the present load of acidity and of the total deposition of sulphur and nitrogen.

Since wet deposition mostly contributes to the total deposition of nitrogen - 70-77% - (see figure C) and to the present load of acidity - 68-80% -, the temporal trends of wet deposition are found to be similar to those of total deposition. As a consequence of reduced sulphur deposition the relative importance of sulphur compounds within the total deposition of acidifying compounds has decreased from 42% to 26% while that of nitrogen compounds has increased from 58% to 74% during the last 20 years.

Figure C *Contribution of wet deposition and dry deposition of gases and aerosols to total acidifying load.*



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