3 Sulphur and nitrogen input-output budgets at ICP Integrated Monitoring sites in Europe in 1990–2012

Progress report

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3.1

Introduction

Detrimental effects of transboundary air pollution led to international agreements to reduce emissions of sulphur and nitrogen in Europe and North America. Due to the implementation of successful emission reduction measures, European emissions of sulphur dioxide (SO_2) have declined by 71% between the years 1990 and 2011 and those of nitrogen (N) compounds by 42% (NO_x) and 31% (NH_3) (Schulz et al. 2013). The emission control programmes have been most successful for SO_2, while nitrogen emissions have stabilized (NO_x) or increased slightly (NH_3) during the 2000s (Amann et al. 2013). The trend analysis of SO_4 concentration for nearly 200 acid-sensitive UNECE ICP Waters monitoring sites in Europe and North America in 1990–2011 indicated that uniform decline of SO_4 concentrations has caused a widespread recovery from acidification of sensitive ecosystems (Garmo et al. 2014). In contrast, only a few

significant trends and regional patterns were found for nitrate (NO_3), but concentrations were decreasing rather than increasing. In the past, nitrogen has played a minor role in acidification, but its relative importance is increasing because N emissions have decreased less than sulphur emissions. Recently, the deposition of reactive nitrogen has been shown to pose a threat to remote terrestrial and aquatic ecosystems through nutrient enrichment (Stevens et al. 2011, Lepori & Keck 2012).

In order to assess the ecosystem benefits of costly emission reduction policies, the importance of long-term integrated environmental monitoring approach including physical, chemical and biological variables is clearly indicated. Mass balance budgets integrate information about the complex chemical and biological recovery processes that govern the retention or release of sulphur and nitrogen compounds and regulate acid production and buffering in both the terrestrial and aquatic portions of catchments in the ecosystem. Long-term assessment of mass balances in hydrologically and geologically well-defined ICP Integrated Monitoring (IM) catchments gives important information for the identification of ecological effects of different anthropogenically derived pollutants, and for the documentation of the effects of emission reduction measures.

The previous IM Annual Report (Vuorenmaa et al. 2013) presented estimates of annual mass balance budgets for sulphur and nitrogen at IM sites for the periods 1990–2010. These results were consistent with budget calculations for a number of other studies from European forested catchments (de Vries et al. 2001, 2003, Prechtel et al. 2001) indicating that forest soils are now releasing stored airborne S that had accumulated in the past.

A new assessment is described in the present report with updated data for the period 1990–2012. This progress report summarizes the main results regarding the calculation of fluxes and trends of sulphur and nitrogen compounds, which will be published in the scientific paper submitted by the end of 2014.

3.2

Materials and methods

Annual input-output budgets for sulphate (SO_4) and total inorganic nitrogen (TIN = NO_3 + NH_4) in the period 1990–2012 were calculated for a selection of 17 IM sites (CZ01, CZ02, DE01, EE02, FI01, FI03, IT01, LT01, LT03, LV01, LV02, NO01, NO02, SE04, SE14, SE15, SE16). In addition, annual output fluxes for organic nitrogen were calculated for 16 sites (CZ02, DE01, EE02, FI01, FI03, IT01, LT01, LT03, LV01, LV02, NO01, NO02, SE04, SE14, SE15, SE16) during the period 1990–2012, according to data availability. For the location of the sites see Fig.1.1 in Chapter 1. The selection of catchments was guided by the availability of deposition (bulk and throughfall) data and surface water chemistry and runoff volume data in the ICP IM database. As IM sites are forested catchments, the dry deposition (gases and particles filtered by the canopy) highly contributes to the total deposition. Total deposition of sulphate (meq m^{-2} yr^{-1}) i.e. the input of wet and dry deposition to the catchment was estimated from bulk deposition (open area) and throughfall (forest stands) measurements. Total deposition of inorganic nitrogen (TIN) was calculated using the same method. Total deposition of N to the forest floor is dependent on wet and dry deposition, and net exchange of material with the vegetation. Because of the strong impact of canopy processes, bulk deposition measurements for TIN were also reported. Annual total deposition fluxes to the catchments were calculated as the sum of monthly values. Output fluxes of TIN (meq m^{-2} yr^{-1}) from the catchments were calculated as the product of measured catchment discharge and ion concentrations. Output fluxes of TIN were calculated as a sum of total inorganic nitrogen (TIN = NO_3 + NH_4) (meq m^{-2} yr^{-1}). Loss of organic

nitrogen was calculated as total nitrogen loss minus TIN loss. Annual runoff water element fluxes were calculated by summing mean monthly fluxes, obtained from monthly mean water flux and monthly mean solute concentration.

At each site trends in fluxes outlined above were analysed using the non-parametric Seasonal Kendall test (Hirsch et al. 1982) applied to annual data. The magnitude of trend was estimated by the Theil-Sen slope estimation method (Sen 1968). The unit of the slope estimate for yearly based data is meq m^{-2} yr^{-1} for fluxes. A statistical significance threshold of $p < 0.05$ was applied to the trend analysis, i.e. providing at least 95 % confidence that the detected trend was significantly different from a zero trend.

In order to quantify the retention or release of sulphur and nitrogen in the catchment, a percent net export (pne) was calculated. The percent net export is defined as: pne = (output–deposition)*100/deposition. Positive pne values indicate release and negative pne values indicate retention in the catchment. For sulphur, total deposition estimate (bulk + throughfall) was used. In the case of nitrogen, bulk deposition in open area was generally larger than throughfall deposition (a surrogate for total deposition) and therefore bulk deposition measurements were used as N deposition estimates in the pne calculations.

3.3

Results and discussion

Large differences in the deposition of sulphur and nitrogen can be observed between the different sites, with the highest values in central Europe and southern Scandinavia and lowest values at sites in northern regions (Fig. 3.1). This reflects well-known gradients in European emissions and deposition of air pollutants. Bulk deposition of TIN (NO_3 + NH_4) generally exceeded SO_4 deposition on an equivalent basis at most of the sites. At most sites, total deposition of N has been lower than that of bulk deposition, indicating canopy uptake. However, at some sites total deposition of N roughly equals with that of bulk deposition, or was higher. This may indicate the importance of dry deposition, but may also indicate the limited ability of forests to store nitrogen.

A statistically significant downward trend ($p < 0.05$) of total sulphur deposition from 1990 to 2012 was observed at all studied IM sites (Table 3.1). As a response to decreased S deposition, sulphate fluxes in runoff have decreased at 15 out of 17 sites, being significant at 50% of the sites. Moreover, a weak decreasing trend ($p < 0.10$) was observed at four sites. Thus, the regional-scale decreases of sulphate deposition and runoff water fluxes observed in the earlier trend assessments (Forsius et al. 2001, Kleemola 2005, Kleemola & Forsius 2006, Vuorenmaa et al. 2009) have continued to decline. Bulk deposition of nitrogen has also decreased at almost all sites (16 out of 17), being significant at 65% of the sites. Total deposition of nitrogen, indicating N fluxes through the canopy to the forest floor, has decreased in 12 sites, being significant at 50% of the sites. Total deposition of N has decreased less (mean annual change -0.70 meq m^{-2} a^{-1}) than that of bulk deposition (mean annual change -1.02 meq m^{-2} a^{-1}). In contrast to sulphate, total inorganic nitrogen (TIN) fluxes in runoff showed mixed response with both decreasing and increasing trends. Statistically significant decreasing trends were observed at five sites and increasing trends at two sites (DE01, Forellenbach, Germany and SE14, Aneboda, Sweden). The significant increasing trends for these two sites are probably due to excess N mineralization and increased NO_3 leaching, resulted from forest damage and dieback in the areas due to storm logging and bark beetle infestation (Beudert et al. 2007, Löfgren et al. 2011).

Sulphate budgets showed increasing percent net exports (pne) at majority of the sites, indicating a net release of previously stored SO_4, particularly during the past 15

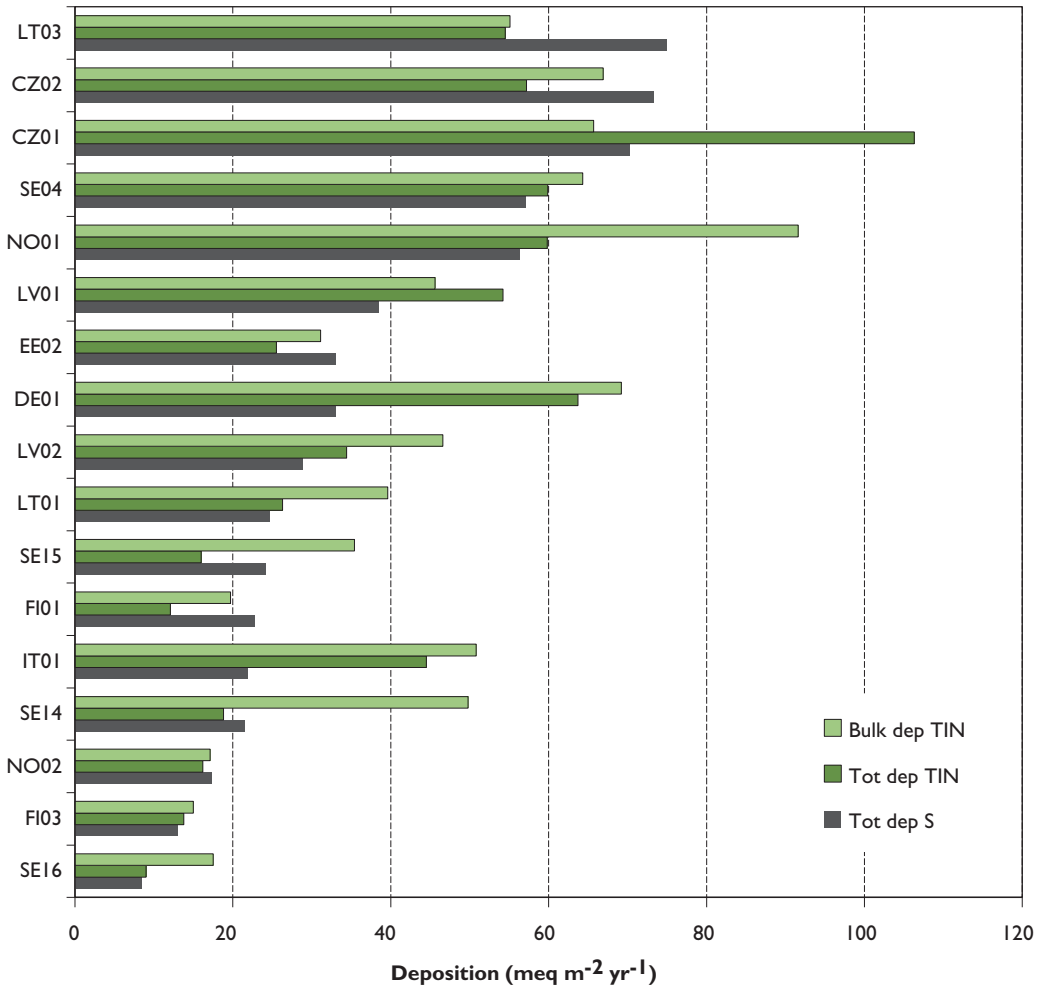

Figure 3.1 Mean annual total deposition of sulphur (SO_4, meq m^{-2} yr^{-1}) and total and bulk deposition of total inorganic nitrogen (TIN = NO_3 + NH_4, meq m^{-2} yr^{-1}) at ICP IM sites in 1995−2012.

years (Fig. 3.2, Table 3.1). This process has taken place both in high and low sulphur deposition areas. For instance, the site CZ02 (Lysina, Czech Republic) has been exposed to high sulphur deposition (> 200 meq m^{-2} a^{-1}) in the early 1990s, but has also been subjected to drastic decrease of S deposition during the 1990s (Table 3.1, Vuorenmaa et al. 2013) and the increase of net release of SO_4. In the low deposition area (< 30 meq m^{-2} a^{-1}) at the site FI03 (Hietajärvi, Finland), sulphate has mainly retained to the catchment, but the net retention rate has decreased over the past 20 years and at present SO_4 input roughly equals with the output. In the selected set of IM sites (Fig. 3.2a), median values for S pne exhibit a significant increase ($p < 0.001$, 3.2 % yr^{-1}) in 1990–2012. A net release of stored SO_4 is considered to act as a H^+ source at many IM sites (Forsius et al. 2005), and SO_4 remains the dominant source of actual soil acidification despite the generally lower input of S than N in European forested ecosystems (deVries et al. 2003). Several processes, including desorption and excess mineralisation, regulate the long-term response of soil S, and a differentiation is necessary for assessing the effects of emission reductions on acidification recovery and for predictions of future responses (Markewitz et al. 1998, Prechtel et al. 2001). In general, many of these S retention/release processes are also sensitive to changes in climatic variables, and would therefore be affected by climate change (e.g. Wright 1998, Benčoková et al. 2011).

Table 3.1 Trends of annual input (deposition) and output (runoff water) fluxes and percent net export (pne) for sulphate (SO_4) and total inorganic nitrogen (TIN = NO_3 + NH_4) at studied IM catchments in 1990–2012. A statistically significant trend ($p < 0.05$) of annual change (meq m^{-2} yr^{-1} for fluxes, % yr^{-1} for pne) is indicated in bold and a potential trend ($p < 0.10$) is indicated in italics.

Catchment	Data	Total SO_4 deposition	SO_4 output	SO_4 pne	TIN bulk deposition	TIN total deposition	TIN output	TIN pne
		(meq m^{-2} yr^{-1})		(% yr^{-1})	(meq m^{-2} yr^{-1})			(% yr^{-1})
LT03, Zemaitija	1996-2012	**-2.35**	1.26	**8.05**	**-1.23**	**-1.11**	0.03	*0.12*
CZ02, Lysina	1990-2012	**-10.05**	**-7.08**	**8.07**	**-1.89**	**-1.49**	**-0.76**	**-0.71**
CZ01, Anenske Povodi	1990-2012	**-6.44**	1.06	**6.30**	**-1.07**	0.90	**-0.15**	**-0.21**
SE04, Gårdsjön	1990-2012	**-3.86**	**-4.46**	1.03	**-1.43**	**-1.92**	0.05	**0.12**
NO01, Birkenes	1990-2012	**-3.45**	**-3.59**	**2.02**	-0.58	**-1.16**	-0.15	-0.11
LV01, Rucava	1993-2009	**-1.48**	-4.43	-1.60	-0.93	*-2.03*	0.10	0.19
EE02, Saarejärve	1995-2012	**-1.90**	-0.66	**7.58**	-0.09	-0.16	0.18	1.02
DE01, Forellenbach	1991-2012	**-2.78**	**-1.08**	**10.15**	**-1.24**	0.31	**2.95**	**5.03**
LV02, Zoseni	1993-2009	**-1.06**	*-5.40*	-7.82	**-4.15**	**-2.56**	**-0.27**	-0.16
LT01, Aukstaitija	1993-2012	**-1.23**	-7.96	6.68	**-1.23**	**-1.42**	-0.11	-0.06
SE15, Kindla	1996-2012	**-1.39**	**-3.62**	-4.35	**-0.79**	**-0.79**	**-0.03**	-0.04
FI01, Valkea-Kotinen	1990-2012	**-1.16**	-0.52	*1.88*	**-0.36**	-0.05	-0.01	0.04
IT01, Renon-Ritten	1995-2012	**-1.68**	0.05	**2.92**	*-0.67*	-0.56	-0.01	0.00
SE14, Aneboda	1996-2012	**-1.58**	**-3.16**	4.06	**-0.91**	0.25	**0.24**	**0.62**
NO02, Kårvatn	1990-2012	**-0.42**	*-0.39*	0.63	0.16	0.15	0.00	-0.10
FI03, Hietajärvi	1990-2012	**-0.58**	**-0.33**	**0.82**	-0.17	0.07	**-0.02**	-0.02
SE16, Gammtratten	1999-2012	**-0.44**	*-0.94*	-0.34	**-0.72**	**-0.30**	*-0.04*	*-0.15*

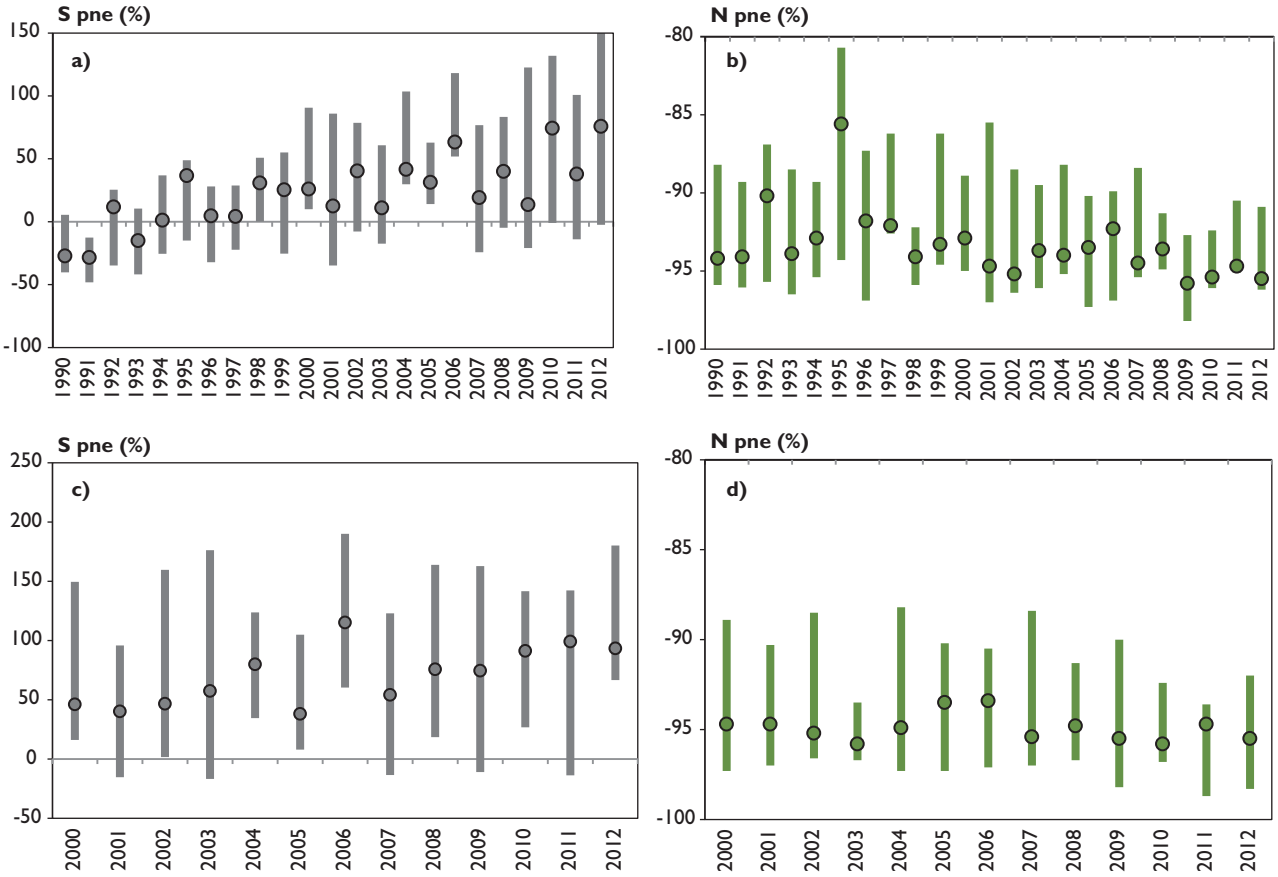

Figure 3.2 Percentiles (25%, median 50%, 75%) of percent net export (pne, %) of sulphate (SO_4) and total inorganic nitrogen (TIN) for the IM sites CZ01, CZ02, DE01, FI01, FI03, NO01, NO02, SE04 in 1990–2012 (a and b, respectively) and for the sites CZ01, CZ02, DE01, EE02, FI01, FI03, IT01, LT01, LT03, LV01, LV02, NO01, NO02, SE04, SE14, SE15, SE16 in 2000–2010 (c and d, respectively). DE01 and SE14 were omitted from the calculation of pne for TIN due to excess N mineralization after Norway spruce (*Picea abies*) dieback due to a bark beetle attack in 1996–1997 and storm logging / bark beetle attack in 2005–2009, respectively.

Nitrogen is generally the growth-limiting nutrient in forest ecosystems, and the uptake of available N compounds is efficient. In contrast to sulphur, nitrogen deposition is usually retained in boreal terrestrial ecosystems; typically < 10% is leached in runoff, mostly as NO3. The percent net export (pne) of nitrogen has generally ranged between –98% and –88% at the studied IM sites during the 2000s (Fig. 3.2b), indicating a strong retention of N in the catchment, and over 50% of the sites exhibit increase in net retention. Correspondingly, median values for TIN pne exhibit a significant decrease ($p < 0.05$, -0.11 % yr^{-1}) i.e. increase in net retention in 1990–2012 (Fig. 3.2b).

Although nitrogen has played a minor role in the acidification in the past, the role of nitrate as an acidifying agent may increase, when continued high nitrogen deposition may result in nitrogen saturation of terrestrial ecosystems, and excess NO_3 leach to surface waters (e.g. Aber et al. 1989, Dise & Wright 1995, Macdonald et al. 2002, Oulehle et al. 2012). Nitrogen saturation may require many decades to occur, at least at levels of N deposition typical for Europe (Wright et al. 2001).

Although the effects of anthropogenic nitrogen inputs on the dynamics of inorganic N in watersheds have been studied extensively, the influence of N enrichment on organic N loss is not as well understood (Pellerin et al. 2006). Studies comparing dissolved organic nitrogen (DON) losses from old-growth forests have reported that DON may account for 60–95% of total dissolved nitrogen losses from minimally disturbed watersheds (Perakis & Hedin 2002, Van Breeman 2002).

Similar results were observed in the IM catchments (Vuorenmaa et al. 2013). Part of this DON may decompose in the downstream surface waters or sea areas (releasing inorganic N compounds) and may thus contribute to detrimental N effects.

In forested watersheds, there is evidence of the effects of nitrogen enrichment on DON production in soils. Plot-scale inorganic N fertilization studies have reported increased DON concentrations in both the forest floor (McDowell et al. 2004) and mineral soils (Pregitzer et al. 2004). The first assessment of the relationship between organic nitrogen loss and N deposition at IM sites gave the signal on link between N deposition and organic nitrogen loss, suggesting that the sites with higher N deposition exhibit also higher organic N loss in runoff (Vuorenmaa et al. 2013). Therefore, DON leaching will receive increasing attention in the forthcoming scientific IM paper. Climate change impacts on mineralization of organic nitrogen and leaching of organic matter, and potential risk for elevated N loss from watersheds to surface waters may be anticipated in the future.

3.4

Conclusions

Forest soils are now releasing sulphur that had accumulated in the past. The more efficient retention of nitrogen than sulphur results in generally higher leaching fluxes of SO_4 than those of NO_3 in European forested ecosystems. Sulphate thus remains the dominant source of actual soil acidification despite the generally lower input of S than N. Organic nitrogen may account for significant fraction of total nitrogen, and N deposition may increase organic N loss in forested catchments. Continued work on processes regulating both N and S retention and release in terrestrial ecosystems is therefore needed. This would be important for assessing the effects of emission reductions on acidification recovery as well as other N pollution problems in semi-natural ecosystems. Many of these S and N retention processes are also sensitive to changes in climatic variables, and would therefore be affected by future climate changes.

The next phase of the work on mass balances of S and N for IM sites will be finalization of a scientific paper, involving assessment of the role of organic nitrogen in mass balance budget. The national focal points and the representatives for the sites will be invited to assist with these activities.

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