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TRANSBOUNDARY AIR POLLUTION

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RECENT RESULTS AND UPDATING OF SCIENTIFIC AND TECHNICAL KNOWLEDGE

ASSESSMENT OF LONG-TERM TRENDS OF DEPOSITION AND SURFACE WATER QUALITY

Report by the Programme Centre of the International Cooperative Programme on Integrated Monitoring of Air Pollution Effects on Ecosystems

I. INTRODUCTION

1. The results of integrated monitoring are presented in this report in accordance with the Convention's 2009 workplan (ECE/EB.AIR/96/Add.2, item 3.6 (c)) approved by the Executive Body at its twenty-sixth session in December 2008.

2. It is anticipated that implementation of the 1999 Gothenburg Protocol to Abate Acidification, Eutrophication and Ground-level Ozone (Gothenburg Protocol) will cut European emissions of sulphur (S) by at least 63 per cent, nitrogen (N) oxides by 41 per cent and ammonium (NH_4) by 17 per cent by the year 2010 as compared to 1990. It is essential that

scientific evidence is available for assessing the success of international emission reduction policy as well as the ecosystem benefits of the large and costly investments.

3. This report summarizes main results of a trend analysis of monthly data on bulk deposition (PC, precipitation chemistry), throughfall deposition (TF) and run-off water (RW) chemistry at 33 sites of International Cooperative Programme on Integrated Monitoring of Air Pollution Effects on Ecosystems (ICP Integrated Monitoring) for the period 1993–2006. It updates an earlier trend assessment for the period 1993–2003 (Kleemola and Forsius 2006). Details can be found in ICP Integrated Monitoring annual report 2009.

II. MATERIALS AND METHODS

4. Monthly concentrations and fluxes for PC, TF and RW or soil water (SW, if no RW data was available) were used in the trend assessment for the individual sites. PC, TF and output (RW) fluxes were calculated from the quality and quantity of water using mean monthly values for water fluxes and chemical analyses.

5. Sites and records were accepted for the statistical analyses, when: (a) data started no later than 1999 and continued at least until 2005; and (b) data were available for at least 7 out of 14 years for the period 1993–2006. Trends were evaluated for non-marine values, where * denotes non-marine fraction, of sulphate (SO_4*), sum of calcium and magnesium (Ca*+Mg*), protons (H^+), nitrate (NO_3) and acid neutralizing capacity (ANC). NH_4 was also included, but the results are not presented due to the limited number of sites with available data. ANC was calculated as the sum of base cations minus the sum of acid ions, i.e. $\Sigma(Ca+Mg+Na+K+NH_4) - \Sigma(SO_4+NO_3+Cl)$ (unit: μeq/l).

6. Thirty-three sites had enough data at least for PC to perform the trend analysis. Twenty-nine sites had sufficient data for TF. From three sites, two separate plots with different dominant species were included in the analysis, which resulted in the total number of 32 TF plots. Twenty-two sites had either *Picea abies* or *Pinus sylvestris* as the dominant species for the whole site or for at least one TF measurement plot. Twenty-seven sites had data for either RW or SW. Average yearly fluxes were also calculated as the average of last five years with available data, generally for the period 2002–2006, in order to reduce yearly variability.

7. Trends in chemical parameters and fluxes were analysed using the non-parametric seasonal Kendall test (SKT) (Hirsch et al. 1982) applied to monthly data at each site. SKT has become a standard method for detecting site-specific trends in water quality data. It can accommodate the non-normality, missing data and seasonality that are common in data of this type. Nevertheless, it is a powerful trend test in a statistical sense (Loftis and Taylor 1989). The magnitude of trend was estimated by the Theil-Sen slope estimation method. This method calculates the median of all between-year differences in the selected variable. The unit of the slope estimate for monthly based data is μeq l^{-1} a^{-1} for concentrations and meq m^{-2} a^{-1} for fluxes. A statistical significance threshold of $p < 0.05$ was applied to the trend analysis. It provides at

least 95 per cent confidence that the detected decreasing or increasing trend was significantly different from a zero trend.

III. MAIN FINDINGS AND CONCLUSIONS

8. Statistically significant downward trends of SO_4 concentrations in PC and TF were observed at most sites. Of 34 sites with sufficient data for trend analysis of PC, all sites had downward trend and 28 sites a statistically significant decrease (figure 1). Of 32 plots, here representing 29 sites, TF concentrations in deposition the corresponding number was 28 plots, here representing 25 different sites. The results showed a clear response to decreasing emissions. The trends for calculated fluxes of SO_4 were similar, but the number of statistically significant results was lower than for the concentration-based assessments. The annual decreases of SO_4 concentrations in both PC and TF for the period 1993–2003 (Kleemola and Forsius 2006) were steeper than for the period 1993–2006. This trend pattern probably correlated with S emissions, as reductions of European sulphur dioxide (SO_2) emissions have slowed down during the recent decade (www.emep.int).

9. Much fewer statistically significant trends were observed for NO_3 than for SO_4 in both PC and TF deposition (figure 1). The overall situation was different with both decreasing and increasing trends detected. For the PC deposition the number of sites with statistically significant decrease in NO_3 concentration was 6 out of 34 sites, while for TF deposition it was 3 sites. Significant increases NO_3 concentrations of PC were not detected. For the estimated NO_3 fluxes of PC, there were two significant decreasing and one increasing trend.

10. The acidity of the deposition showed decreasing trends on the regional scale. The number of sites with decreasing H^+ concentrations of PC was 19 and 14 for H^+ flux. One site showed a significant increase in both PC concentration and deposition flux. For H^+ concentrations of TF, the respective numbers were 17 plots representing 16 sites, and 16 plots representing 15 sites for H+ fluxes. No site showed an increase in H^+ concentrations and fluxes in TF. The TF deposition reflected better the total load to the ecosystem, as it accounted for changes in dry deposition fraction as well. The observed increasing trends in H^+ concentrations were partly explained by a decrease in the neutralizing base cation deposition, which has affected the acidity load despite the regional-scale decrease in SO_4.

11. The results concerning RW/SW showed a more mixed response than in deposition. This is due to the complex interaction of soil and catchment processes affecting the concentrations and fluxes of the different ions. Of the 27 sites with the available concentration data, statistically significant decreasing SO_4 trends were observed at 18 sites and an increasing trend at one site (figure 2). Due to missing data on soil or RW fluxes, flux estimates could be made only at 19 sites. Seven sites showed a significant decrease in SO_4 flux, and one site an increase. Previous input-output budget studies for forested sites in Europe, including ICP Integrated Monitoring sites, have often showed a net release of SO_4. This indicated that forest soils were now releasing

S, which had accumulated in the past (de Vries et al. 2001 and 2003, Prechtel et al. 2001, Forsius et al. 2005). As a general trend, the sites were thus responding to the decreasing S deposition.

12. NO_3 concentrations in RW/SW showed mixed response with both decreasing and increasing trends (figure 2). Statistically, significant decreasing trends were observed at six sites and increasing trend at one site. Respective numbers for NO_3 fluxes were four with decreasing and five with increasing trends. Few significant temporal trends have generally been observed regarding annual NO_3 concentrations in RW in background areas in Europe (Forsius et al. 2001, Wright et al. 2001).

13. ANC at acid-sensitive sites in northern Europe showed increasing trends, thus indicating recovery from acidification. More significant trends in concentrations were observed for ANC, increasing at 13 out of 27 sites, than for H^+, which is related to pH, decreasing at 6 out of 26 sites. ANC is generally a more stable indicator and less affected by short-term processes. Many of these sites were not sensitive to acidifying deposition (e.g. Forsius et al. 2005); therefore, large changes in acidity variables would not be expected. The increasing ANC trends for these acid-sensitive sites indicated recovery from acidification and thus a positive response to the decreasing emissions.

14. The new results of the ICP Integrated Monitoring sites confirmed previously observed regional-scale decreasing trends of S in deposition and RW/SW. Acid-sensitive sites in northern Europe indicated continuing recovery from acidification. The situation on N was quite different, with few decreasing trends in deposition and both decreasing and increasing trends in RW/SW. Critical load calculations for Europe also indicate exceedance of the N critical loads over large areas. N thus requires continued attention as an air pollutant in Europe.

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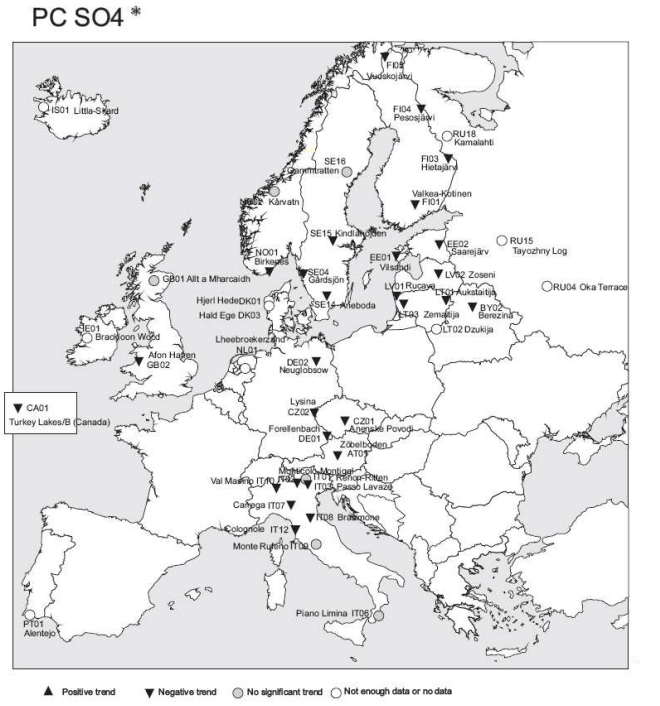
Figure 1. Increasing and decreasing slopes (triangles indicate the direction) for statistically significant ($p<0.05$) trends for SO_4 and NO_3 concentrations in PC



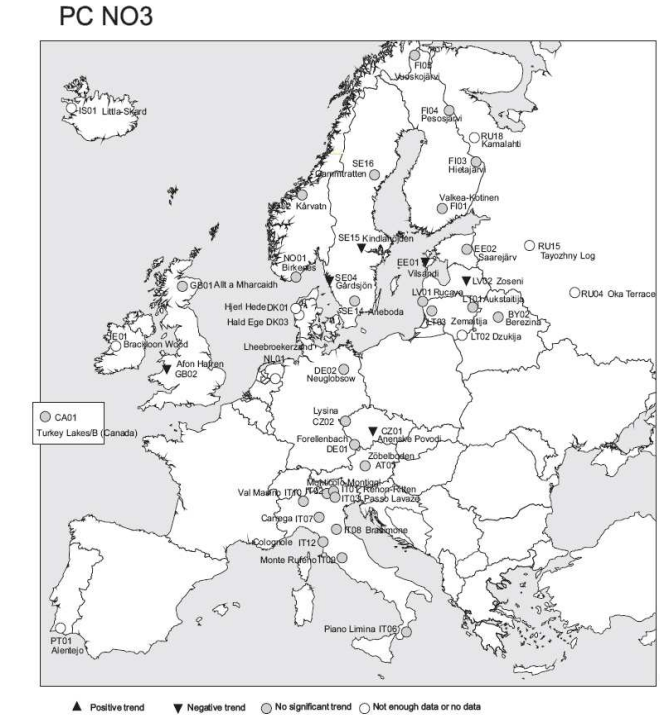


Figure 2. Increasing and decreasing slopes (triangles indicating the direction) for statistically significant ($p<0.05$) trends for SO_4 and NO_3 concentrations in RW or SW chemistry