Tellus (2000), 52B, 60–73
Printed in UK. All rights reserved

TELLUS
ISSN 0280–6509

Atmospheric concentrations and deposition of oxidised sulfur and nitrogen species at Petaling Jaya, Malaysia, 1993–1998

By G. P. AYERS[1]*, LEONG CHOW PENG[2], LIM SZE FOOK[2], CHEAH WAI KONG[3], R. W. GILLETT[1] and P. C. MANINS[1], [1]*CSIRO Atmospheric Research, PMB 1, Aspendale, Vic. 3195, Australia;* [2]*Malaysian Meteorological Service, Jalan Sultan, 46667 Petaling Jaya, Malaysia;* [3]*Tenaga Nasional R&D Berhad, PO Box No 80, 43007 Kajang, Selangor, Malaysia*

(Manuscript received 10 February 1999; in final form 27 May 1999)

ABSTRACT

Wet-only rainwater composition, acid-precursor gas mixing ratios and aerosol loading were determined from weekly-averaged samples at Petaling Jaya, Malaysia, over the five year period from March 1993 to March 1998. Annual deposition fluxes of acidic sulfur and nitrogen species estimated from these data show this site to be heavily impacted by acidic deposition, with total oxidised sulfur plus nitrogen deposition in the range 277–480 meq m^{-2} yr^{-1}. Average contributions were 56% as sulfur species, 44% as nitrogen species, with wet deposition in this region of high rainfall accounting for 67% of total deposition. Thus total acid deposition fluxes were equivalent to levels that provided motivation for emissions reduction programs in both Europe and North America. The possibility of adverse environmental effects in Malaysia caused by acid deposition therefore merits serious consideration and assessment.

1. Introduction

Concern over adverse environmental effects of acid deposition figures heavily in the justifications given in Europe and north America for emissions reductions programs such as the European sulfur and nitrogen oxides Protocols and Title IV of the Clean Air Act Amendments in the US (Barrett et al., 1995; NAPAP, 1991; EPA, 1995 and references therein). Reductions in sulfur emissions and reductions or stabilisation of nitrogen oxides emissions have followed these initiatives in Europe and north America, stimulating a broadening of focus to consider other parts of the globe where acid precursor emissions continue to rise rapidly in concert with economic development, such as eastern and southern Asia, southern Africa and South America (Rodhe et al., 1995a, b; Galloway, 1995).

Here we consider one of these regions, the Asian region, which has been identified as potentially subject to excessive atmospheric acidity and acid deposition (Galloway, 1988a,b; McDowell, 1988; Rodhe and Hererra, 1988; Foell and Green, 1991; Ayers, 1991; Kato and Akimoto, 1992; Rodhe et al., 1992; Carmichael et al., 1995). Specifically, we have carried out a 5-year study of atmospheric acidity and acidic deposition at an equatorial location, Petaling Jaya (PJ), in Malaysia. Our aim was to document levels of atmospheric acidity in an Asian region containing significant acid precursor emissions, and to discuss the measured acid levels within the context provided by similar data from European and north American assessments of acid deposition and implications for the

* Corresponding author.
e-mail: chief@dar.csiro.au.

environment. A preliminary report on rainwater composition in this region (MMS, 1988) had suggested a decade ago that rainwater in this region was acidified significantly by nitric and sulfuric acidity derived from anthropogenic NO_x and SO_2 emissions.

2. The Petaling Jaya site

PJ is located within the Klang Valley (Fig. 1), which covers an area of approximately 3000 km^2 and stretches approximately 80 km inland in a north-easterly direction from the west coast of peninsular Malaysia to the foothills of the Genting Highlands. The Klang Valley region had a population of 2.95 million people in 1991 (August 1991 census), and contains a broad mix of urban through to light and heavy industrial activities. One large fossil-fuel-fired power plant is located at the western (coastal) end of the Klang Valley at Meru, and another in Klang, with the central business district of the national capital, Kuala Lumpur, located at the eastern end. PJ is located approximately mid-way along the Valley. In 1992 Klang Valley emissions of NO_x and SO_2 were reported to be 54 ktonnes (as NO_2) and 36 ktonnes respectively, with 67% of NO_x emissions attributed to mobile sources and 86% of SO_2 emissions attributed to "factories" (JICA, 1993). While not large emissions by world standards, build-up of emissions in the regional atmosphere is common because of poor advective transport resulting from the prevalence of calm and low wind-speed conditions in the region. Annual average wind speeds at six sites in the Klang Valley region have been reported to lie between 0.5 and 1.3 m s^{-1} (JICA, 1993).

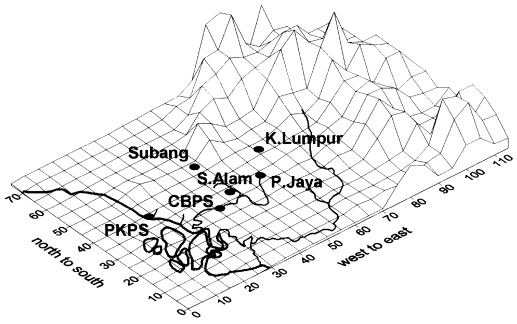

Fig. 1. Map of Klang Valley region showing locations of Kuala Lumpur (Lat. 3°08′N, Long. 101°42′E), the industrial region of Shah Alam, location of the Meteorological Service sites at Subang and PJ, and locations of the major thermal power stations (PKPS and CBPS). The terrain rises to peaks over 1000 m in the Genting Highlands to the Northeast of Kuala Lumpur.

The measurements reported here were made at the headquarters of the Malaysian Meteorological Service (Perkidhmatan Kajicuaca Malaysia) at PJ. This is located in a regional commercial precinct that includes other government department buildings, shopping strips, hotels and other low-rise commercial buildings. The site is approximately 200 m from the nearest major roadway.

3. Experimental

The sampling equipment consisted of a solar-powered wet-only rainwater sampler (Ecotech Model 200), a Ferm-type passive gas sampler (Ferm, 1991; Ayers et al., 1998a) for the gases SO_2 and HNO_3 with a separate passive gas sampler for NO_2, and a custom-built low-volume aerosol sampler with an inlet configuration chosen to approximate the PM10 urban aerosol standard (50% cut-point at 10 μm aerodynamic diameter). The aerosol filter substrate was 1 μm Fluoropore, 47 mm in diameter; average sample volume was 28 m^3.

All samples were taken approximately 1.5 m above ground, with the sampling equipment placed in the grassed compound adjacent to the headquarters building in which local surface meteorological observations are taken. Samples were accumulated for 7 days, with sample retrieval each Tuesday at 0800. The rainwater samples were preserved from biological degradation by placing thymol in the polyethylene rainwater sample bottle prior to installation of the sample bottle in the wet-only sampler (Gillett and Ayers, 1991; Ayers et al., 1998b). The sample bottle was removed from the sampler and shipped to CSIRO with the passive gas samplers and aerosol filters for analysis, ensuring that the rainwater sample did not contact any surface other than the original sample bottle and was always in contact with thymol. Rainwater volume was determined gravimetrically, both from the wet-only sampler and separately from a parallel national-standard

raingauge. Average time between collection of samples and subsequent analysis was approximately 2 months.

Chemical analysis was primarily by Ion Chromatography (Dionex Model DX500), using AS11 anion separation columns and CS12 cation separation columns for both the rainwater and aerosol samples (the latter extracted into 20 ml of high purity water in polyethylene bags). Conductivity was determined by standard electrode methods, as was pH using an Orion Ross electrode and Orion Low Ionic Strength buffers. Aerosol mass loading was determined gravimetrically using a Mettler MT5 microbalance, with filters weighed after 24 hours of desiccation over silica gel.

The SO_2/HNO_3 passive samples were analysed also by IC, using a Dionex AS4A separator column. The NO_2 passive samples were analysed colorimetrically, as described by Ferm (1991).

A variety of data quality assessments suggest that the data are of high quality. For example the ion balance results from the rainwater and aerosol samples are shown in Fig. 2 with the US EPA reanalysis criteria (EPA, 1994). The relatively few points that fall outside the EPA limits are all accepted as valid, since reanalysis did not alter the ion sum values significantly.

In the case of the low-volume aerosol sampler deployed to collect the PM10 aerosol fraction, the gravimetric results were checked against results obtained independently by the Malaysian Meteorological Service using a conventional PM10 high-volume aerosol sampler (Anderson Hivol at 1.1 m^3 min^{-1}). Fig. 3 shows the comparison over a 6 month period in 1994, which shows reasonable agreement between the low and high volume samplers. The data record was essentially complete, with only 2 weekly aerosol samples lost through the study period due to equipment failure.

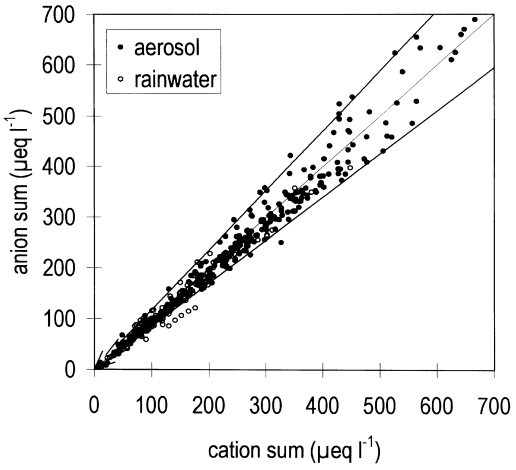

Fig. 2. Ion balance plot for rainwater and aqueous aerosol extracts from PJ. The lines depict the US EPA reanalysis criteria referred to in the text (EPA, 1994).

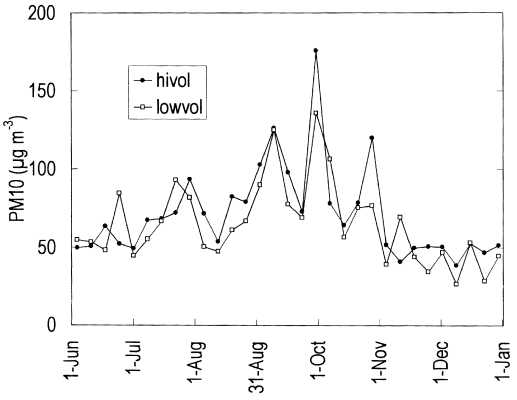

Fig. 3. Comparison of aerosol mass determinations from the MMS PM10 high-volume aerosol sampler and the CSIRO low-volume aerosol sampler for the latter half of 1994.

For the passive gas samplers we rely on the validations by Ferm (1991) in Sweden and Ayers et al. (1998a) in Australia showing that these samplers return unbiased absolute concentration data. Additionally, the samplers were always exposed in duplicate, as recommended by Ferm (1991), so as to provide information on measurement precision. The overall mean deviation between pairs of samplers (221 pairs of each type) expressed as a percentage of the mean of each pair, was 22% for HNO_3, 19% for SO_2 and 9% for NO_2.

4. Results

Illustrative time series results from the different measurement systems are displayed in Fig. 4, which presents data for SO_2 and HNO_3, Fig. 5 which presents the gaseous NO_2 and PM10

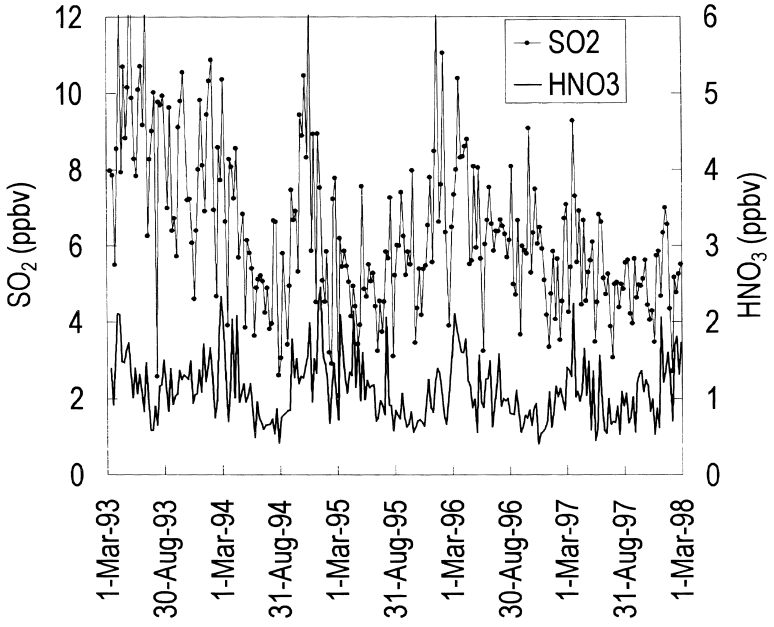

Fig. 4. Time series of gaseous SO_2 and HNO_3 mixing ratios, March 1993–March 1998. Data points are 7 day means.

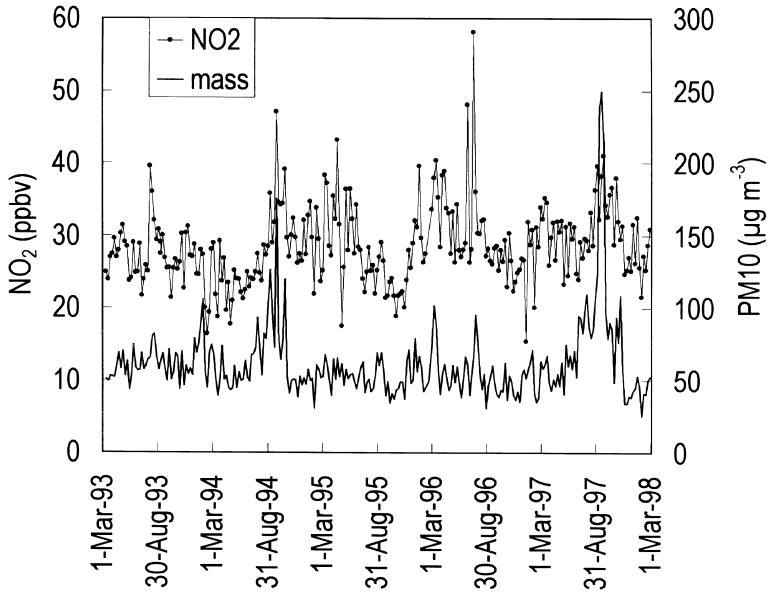

Fig. 5. Time series of gaseous NO_2 mixing ratio and PM10 aerosol mass concentration, March 1993–March 1998. Data points are 7 day means.

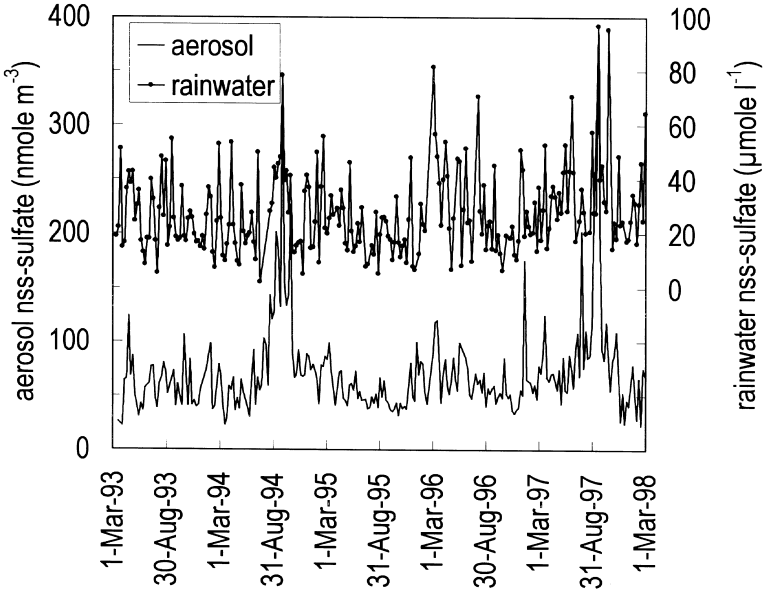

Fig. 6. Time series of aerosol nss-SO_4^{2-} concentration (nmole m^{-3}) and rainwater nss-SO_4^{2-} concentration (μmole m^{-3}), March 1993–March 1998. Data points are 7 day means.

aerosol data, and Fig. 6 which shows the time series for rainwater and aerosol non-sea salt sulfate (nss-SO_4^{2-}). Note in Fig. 5, evidence of the major regional haze events of August–September 1994 and 1997.

Since the focus here is on acid deposition, which is typically discussed in terms of annual averages, the complete dataset on gas, aerosol and rainwater composition is summarised in Tables 1–3 in terms of 5 annual means, with the annual periods ending in March, the month in which the measurements commenced in 1993. The high rainfall in the final annual period is not at odds with the drought conditions that contributed to the haze conditions late in 1997, as it resulted mainly from unusually heavy rainfall in March 1997. Aerosol and rainwater data on organic acids are included in the Tables to complete the picture of ionic composition. However, these acids do not contribute to environmental acidification, being subject to natural processes of biological degradation as neutral species (Herlihy et al., 1987).

Table 1. *Annual mean acidic gas mixing ratios at PJ*

Sample period	Annual mean (ppbv)		
	NO_2	SO_2	HNO_3
9 March 1993–8 March 1994	27.0	8.5	1.3
8 March 1994–7 March 1995	27.5	6.0	1.1
7 March 1995–5 March 1996	27.8	5.7	1.1
5 March 1996–4 March 1997	30.1	6.3	1.0
4 March 1997–3 March 1998	30.3	5.2	1.1

5. Discussion

5.1. Deposition estimation

We concentrate on the nitrogen and sulfur species that dominate acid deposition of anthropogenic origin, and first convert the atmospheric measurements from mixing ratio or concentration data to fluxes. For the rainwater species this is trivial, wet deposition being calculated simply from the product of annual volume-weighted mean concentration and annual rainfall. We consider the annual deposition fluxes to be representative and free from significant bias as in each year the wet-only rainwater collections comfortably exceeded to World Meteorological Organization's criterion that the wet-only sampler collect a minimum of 80% of annual rainfall reported by the

Table 2. *Annual mean concentrations of PM10 aerosol mass and soluble aerosol components at PJ*

	PM10	H^+	Na^+	NH_4^+	K^+	Mg^{2+}	Ca^{2+}	Cl^-	Br^-	NO_3^-	PO_4^{3-}	SO_4^{2-}	$C_2O_4^{2-}$	Formic	Acetic	MSA
year 1	62.1	10.3	13.6	73.5	13.1	3.7	18.8	4.1	0.41	10.3	0.39	60.9	4.1	2.3	3.2	0.19
year 2	62.9	18.0	15.9	116.8	17.0	3.4	14.2	4.1	0.18	12.4	1.18	88.1	7.7	2.7	0.7	0.30
year 3	53.1	14.0	11.9	83.9	12.5	2.3	11.3	2.3	0.06	5.6	0.59	55.2	8.0	1.4	0.3	0.28
year 4	52.0	14.8	9.1	97.7	13.0	1.6	8.0	0.9	0.04	3.2	0.66	64.4	4.7	1.0	0.4	0.25
year 5	75.0	19.1	12.4	149.9	15.1	2.4	11.9	2.4	0.01	13.3	1.41	91.0	7.7	1.5	1.3	0.43

PM10 loading is given in $\mu g\, m^{-3}$, with all other components in $nmole\, m^{-3}$. The annual periods correspond to the data given in Table 1.

Table 3. *Annual mean rainfall and volume-weighed mean rainwater concentrations for ionic components at PJ*

	Rain	pH	H^+	Na^+	NH_4^+	K^+	Mg^{2+}	Ca^{2+}	Cl^-	NO_3^-	PO_4^{3-}	SO_4^{2-}	$C_2O_4^{2-}$	Formic	Acetic	MSA
year 1	3242	4.33	46.8	3.8	16.8	1.2	1.4	6.6	5.8	20.0	0.05	23.4	0.8	8.8	7.2	0.43
year 2	2626	4.30	50.0	5.2	25.8	1.6	1.6	7.2	7.7	23.3	0.19	24.6	1.2	12.1	10.4	0.08
year 3	3030	4.43	36.8	3.6	16.8	1.5	1.4	6.2	5.5	18.7	0.19	18.5	0.9	2.0	2.4	0.12
year 4	3224	4.28	52.6	4.1	19.6	1.4	1.6	7.5	7.5	25.0	0.27	24.1	0.7	5.8	5.7	0.30
year 5	3684	4.19	63.8	4.9	32.1	2.3	1.8	8.1	8.0	32.6	0.20	33.7	0.9	4.1	4.6	0.19

Rainfall is given in mm, with all other components except pH in units of $\mu mole\, l^{-1}$. MSA is methansulfonate. The annual periods correspond to the data given in Table 1.

national raingauge at the sampling site (WMO, 1992).

However, in the cases of gaseous and aerosol species the estimation of dry deposition flux from measured near-surface concentration or mixing ratio data is not straightforward, since the necessary transfer coefficient, the deposition velocity (V_d) is infrequently amenable to measurement and is difficult to characterise theoretically. Here we adopt the semi-empirical, but well documented and widely used "inferential method" as a means of estimating in a generally accepted way values for the deposition velocities of SO_2, NO_2 and HNO_3 at PJ. Briefly, according to the inferential approach the deposition flux, F_d, to the surface of a gas such as sulfur dioxide may be described in terms of a dry deposition velocity, V_d:

$$F_d = CV_d = C/R_t = C/[r_a + r_b + r_c], \quad (1)$$

where C is the near-surface concentration of the relevant trace gas. V_d is inversely proportional to the sum of 3 resistance terms:

$$V_d = (r_a + r_b + r_c)^{-1}. \quad (2)$$

These terms characterise the critical factors controlling surface-atmosphere exchange (Hicks et al., 1987), where r_a is aerodynamic resistance, r_b is surface boundary layer resistance, and r_c is surface or canopy resistance. Methodologies for estimating the resistance terms from meteorological parameters and surface properties were taken from Hicks et al. (1987), Baldocchi et al. (1987), Schlünzen and Pahl (1992), Walcek et al. (1986), Erisman et al (1993), and Wesely et al. (1983). The procedures were implemented in a manner similar to that of Hicks and Meyers (1991) in the computer code DryDep2. Deposition velocities were computed for all hours of the year, employing a meteorological file of all relevant parameters at hourly intervals, available from PJ for calendar year 1992.

The overall average deposition velocity for SO_2 at PJ is estimated to be 0.0031 m s^{-1}. This value is practically the same as employed by Ayers and Yeung (1996), based on literature survey information, in a study of acid deposition in Hong Kong. However when effects of covariance between the diurnal cycles in deposition velocity and SO_2 concentration are considered, that is, if annual deposition is to be computed from the product of annual average SO_2 concentration and average annual deposition velocity, rather from an the annual mean of the hourly products of SO_2 concentration and deposition velocity, this value must be reduced by 25% to 0.0023 m s^{-1}. Here we employed unpublished data from DoE on mean diurnal SO_2 cycles in the Klang Valley to take this covariance into account.

For NO_2, the average deposition velocity at PJ was estimated to be 0.0034 m s^{-1}. In this case, effect of covariance between the diurnal patterns in deposition velocity and gas concentration were larger than for SO_2, leading to an annual mean deposition velocity of 0.0018 m s^{-1} when applied to annual mean NO_2 data.

For HNO_3 the dry deposition velocity is controlled by the aerodynamic and boundary layer resistances alone, and we compute an overall an annual average deposition velocity of 0.0083 m s^{-1}.

For aerosol SO_4^{2-} and NO_3^- an annual deposition velocity of 0.00029 m s^{-1} was estimated for both species.

Uncertainty in the deposition velocity estimates is hard to quantify, however based on discussions in the primary references cited earlier and the results of application of this methodology in the US (Hicks et al., 1991; Meyers et al., 1991) the accumulated annual average values should be at least correct to within a factor of 2.

5.2. *Deposition fluxes*

Wet and dry deposition fluxes of sulfur and nitrogen species estimated from the atmospheric data in Table 1–3, the annual rainfall totals, and the dry deposition velocities estimated as described above, are presented in Table 4. From the data in Table 4, averaging over all NO_x and SO_x species across all five years, the total deposition of oxidised N and S species is found to be 56% as S species, 44% as N species, and 67% wet deposition, 33% dry deposition.

5.3. *Comparison with deposition fluxes from other locations*

Two differing perspectives on the deposition fluxes tabulated in Table 4 may be obtained by comparing these data with data from an unpolluted location on the one hand, and data from regions known to be affected adversely by acid

Table 4. *Annual wet and dry deposition fluxes at PJ (meq m^{-2} yr^{-1})*

	Rainwater			Aerosol			Gas			Total
Sample period	NH_4^+	NO_3^-	nss-SO_4^{2-}	NH_4^+	NO_3^-	nss-SO_4^{2-}	HNO_3	NO_2	SO_2	$NO_x + SO_x$
9 March 1993 –8 March 1994	54	65	150	1	<1	1	14	62	50	343
8 March 1994 –7 March 1995	67	62	128	1	<1	2	12	63	36	304
7 March 1995 –5 March 1996	51	57	111	1	<1	1	11	64	34	279
5 March 1996 –4 March 1997	63	81	154	1	<1	1	11	69	37	354
4 March 1997 –3 March 1998	119	121	246	1	<1	2	12	70	31	483

deposition, on the other hand. Data for an unpolluted tropical location are available from the Northern Territory (NT) of Australia. Like peninsular Malaysia, this is a tropical region. For example, the capital, Darwin, has a mean annual maximum temperature of 31.9°C, mean annual daily minimum temperature of 23.2°C, and is subject to significant annual rainfall (1666 mm; 55 year average). However unlike the Klang Valley region in Malaysia, the NT is relatively unpopulated, with few anthropogenic sources of NO_x and SO_2. Relevant statistics for the NT are a land area of 1.35 million km^2, holding a population of only 170,000 about half of whom reside in the capital city, Darwin. Thus the average population density outside Darwin is only 0.06 km^{-2}. As outlined by Ayers and Gillett (1988) anthropogenic atmospheric emissions in this part of Australia are extremely low by world standards, leading to rainwater composition very low in ionic constituents of anthropogenic origin, especially nitrate and sulfate (Likens et al., 1987; Noller et al., 1990; Gillett et al., 1990; Ayers et al., 1993). We therefore adopt the multi-year rainwater composition data available from the NT site of Katherine (14°28′S, 132°18′E) as representing unpolluted tropical rainwater (Likens et al., 1987).

Data on the gaseous species that act as major contributors to acid deposition at PJ (Table 1) are not available in the NT, where no air quality measurements are made. However, measurements made by the authors with passive samplers identical to those used at PJ have been carried out since June 1993 at Charles Point on the Cox Peninsular, approximately 20 km NW of Darwin. This site was chosen specifically to act as a regional "background" atmospheric chemistry site, away from local anthropogenic emission sources, so well serves our purpose here.

Table 5 contains mean rainwater composition data for Katherine from Likens et al. (1987) for the 5-year period from 1980–1984, and mean gas concentrations from Charles Point averaged over the four years from 1993 to 1997. The Table also contains annual deposition fluxes for N and S species calculated from these data. The comparison between deposition fluxes in Tables 4 and 5

Table 5. *Volume-weighted mean rainwater concentrations and annual average wet deposition fluxes of nitrate and sulfate at Katherine (1980–1984, Likens et al., 1987) and mean passive gas data from Charles Point near Darwin (June 1993–June 1997); annual dry deposition fluxes of the gases were calculated assuming dry deposition velocities as for PJ (see text)*

Rainwater	NH_4^+	NO_3^-	SO_4^{2-}
concentration (μmol l^{-1})	2.9	4.1	2.0
deposition (meq m^{-2} yr^{-1})	3.0	4.3	4.4
Gas	HNO_3	NO_2	SO_2
mixing ratio (ppbv)	0.34	0.75	0.91
deposition (meq m^{-2} yr^{-1})	3.7	1.7	5.4
Total			
total deposition ($NO_x + SO_x$) (meq m^{-2} yr^{-1})		19.5	

is stark: total NO_x plus SO_x deposition of order 300 meq m^{-2} yr^{-1} at PJ is 15× higher that the ~20 meq m^{-2} yr^{-1} estimated from the Northern Territory data.

The second perspective against which to set the PJ data is provided by regions acknowledged for more than two decades to have been impacted adversely by acidic deposition: parts of Europe and North America. Comprehensive descriptions of the vast amount of research undertaken in these regions have been published over the years (Overrein et al., 1980; NAPAP, 1991; RMCC 1992; Hettelingh et al., 1991; Chadwick and Hutton, 1991), and do not require repetition here. For our purposes a convenient recent summary of this work is provided by the World Meteorological Organisation in the form of a Global Acid Deposition Assessment (WMO, 1997). This review shows clearly that in the extensive regions of Europe and North America where anthropogenic emissions dominate the regional atmospheric N and S cycles (i.e., emissions fluxes) by an order of magnitude or more, the concomitant regional deposition fluxes of NO_x and SO_x each typically fall in the range 50–100 meq m^{-2} yr^{-1}, with values double these not uncommon in specific locations (WMO, 1997). Thus total NO_x plus SO_x annual deposition fluxes in the range 200–400 meq m^{-2} yr^{-1} define those regions in Europe and North America in which adverse environmental consequences have been clear (NAPAP, 1991; Hettelingh et al., 1991; Chadwick and Hutton, 1991; RMCC, 1992). Viewed against this background, we can conclude that the acid deposition fluxes determined at the PJ site (Table 4) are equivalent to the levels that have elsewhere motivated anthropogenic sulfur and nitrogen emissions reductions programs (*e.g.* the Sulfur Protocols in Europe and the Title IV amendments in the US). The potential for adverse environmental consequences in the vicinity of PJ therefore must be recognised.

5.4. *Deposition fluxes and critical loads*

While the anthropogenic elevation of the acid deposition fluxes at PJ is unmistakable when these are contrasted with the fluxes in tropical Australia (Table 5) and acidified regions of the northern mid-latitudes (WMO, 1997), elevated acid fluxes alone do not guarantee effects on terrestrial surface water or soil ecosystems. It is the balance between acid input at the surface and the acid buffering capacity of regional soils and surface waters that determines the extent, if any, of adverse effects on the environment. In Europe this balance is expressed in a pragmatic way in terms of regional "critical loads", defined as "a quantitative estimate of an exposure to one or more pollutants below which significant harmful effect on specified elements of the environment do not occur according to our current knowledge" (Nilsson and Grennfelt, 1988). In the case of acidic deposition, the critical load for a given location is specified as an annual total acid deposition flux not to be exceeded (Chadwick and Hutton, 1991; Hettelingh et al., 1991; Hornung et al., 1995). A variety of tractable methods for estimation of critical loads has been developed and applied in Europe (Hettelingh et al., 1991; Chadwick and Hutton, 1991; Downing et al., 1991; Hornung et al., 1995). A practical development of this concept is that of a "target load", which provides flexibility for policy-makers, may be based on a cost-benefit analysis, and does not aim to protect 100% of acid deposition receptors in a given region (e.g., see comments in Ch. 8 of Chadwick and Hutton, 1991).

While the European terminology and the general approach based on critical loads and target loads has also been adopted in Canada (RMCC, 1992), it has not been adopted in the US (EPA, 1995). However such a framework has been under development for the Asian region in recent years (Kuylenstierna et al., 1995; Hettelingh et al., 1995), and while not yet developed to the level of detail available for Europe, the RAINS-Asia integrated acid deposition assessment system (Amann and Dhoondia, 1994) does provide the only published maps of critical loads for the SE Asian region.

The critical loads module of the RAINS-Asia modelling system determines critical loads according to the steady state mass balance approach (Hettelingh et al., 1991; Downing et al., 1991) at relatively coarse resolution ($1^\circ \times 1^\circ$ latitude/longitude). It allows specification of target loads, rather than critical loads, at a given level of receptor protection. Discussion of the methods employed and assumptions involved is provided in the supporting documentation (Amann and Dhoondia, 1994).

Here we have employed the suggestion of Hettelingh et al. (1995) for a target load set to

protect 75% of receptors in the $1^\circ \times 1^\circ$ grid square that covers the Klang Valley region in Malaysia in which PJ and Kuala Lumpur are located. The RAINS-Asia model yielded an acid deposition target load of 33 meq m^{-2} yr^{-1} for this grid square. This relatively low value reflects factors such as the soil types in the region, and especially the high rainfall that defines the regional soil types as highly leached. However it is still at or above the values that apply for the most acid-sensitive regions of Europe where acidification effects have been noted, for example parts of Scandinavia (Chadwick and Hutton, 1991; Hettelingh et al., 1991; Downing et al., 1993). It is also well above the total annual NO_x plus SO_x deposition level of $\sim$20 meq m^{-2} yr^{-1} estimated for the unpolluted Northern Territory of Australia (Table 5), suggesting that if the tropical Australian deposition were representative of "natural" (as opposed to anthropogenic) acid deposition cycles throughout SE Asia, the critical loads in the Klang Valley would not be exceeded by "natural" deposition fluxes.

However the target load of 33 meq m^{-2} yr^{-1} is an order of magnitude below the total annual NO_x plus SO_x deposition levels for PJ given in Table 4. This comparison is essentially unchanged even if allowance is made for the input of base cations that accompanies the nitrate and sulfate deposition in the aerosol or rainwater (Tables 2, 3), since (1) the base cation levels are very much smaller than the nitrate and sulfate levels, and (2) this small base cation input may well be balanced by additional nitrate acidity generated in the soil biologically, following consumption of the high levels of ammonium also deposited in the aerosol and rainwater deposition (see Ch. 3 in Chadwick and Hutton, 1991). This 10-fold excess of deposition over the estimated target load provides a clear imperative for evaluation of potential environmental consequences in the Klang valley region.

6. Uncertainties and open questions

Given the magnitude of the apparent mismatch between acid deposition fluxes at PJ and the regional critical loads it is important to acknowledge the considerable limitations of the present analysis. First, the accuracy of, and uncertainty in, the target loads computed by the RAINS-Asia system have not been specified (Amann and Dhoondia, 1994; Hettelingh et al., 1995), and it is acknowledged that "extensive validation procedures are required to improve the assessment of risk of damage on local scales" (Hettelingh et al., 1995). Moreover, there remains scientific debate about the fundamental validity of the critical loads approach, including questions concerning whether soluble Aluminium in soil solution is the main cause of forest damage, and our ability to model accurately the effects of acid deposition (Løkke et al., 1996; Müller-Edzards et al., 1997; Zhao and Seip, 1991; Mulder and Stein, 1994). Therefore at this stage knowledge of critical loads in the Klang Valley region should be considered to be qualitative at best. Clearly, there is a strong imperative for direct experimental validation of the broad-scale ($1^\circ \times 1^\circ$ resolution) target load, and assessment of its applicability at the PJ site.

Second, the geographic representativeness of elevated levels of acid deposition in the Klang Valley is unknown. The PJ site was chosen partly because it sits roughly in the middle of the industrialised Klang Valley region, so can be expected to reflect a worst-case situation. Given the low wind speeds mentioned earlier, advective transport of Klang Valley emissions to adjacent regions may not be efficient. Therefore a second priority is measurement of acid deposition fluxes at other locations in and outside the Klang Valley, to define the geographic extent of the phenomenon.

Third, the possibility of up to a factor of 2 uncertainty in dry deposition velocities must be acknowledged. This uncertainty is not, however, sufficient to influence the general conclusions concerning overall acid deposition fluxes, since as was noted earlier dry deposition was estimated to account for only one third of the total flux.

In terms of other open questions, it would appear prudent in the face of significant changes in emission sources to maintain ongoing measurements at PJ in order to track the effects of changing emissions on atmospheric levels and deposition fluxes of acidic species. In recent years automobile registrations in Malaysia have been increasing rapidly, for example by 9% from 1994 to 1995 (DoE, 1996a). Concomitant increases have been estimated in NO_x emissions from mobile sources, to the point where by 1995 mobile sources were reported to be the major anthropogenic NO_x

source in Malaysia (DoE, 1996b). It appears noteworthy in this context that the annual mean level of NO_2 measured at PJ rose from 27.0 ppbv in 1993/4 to 30.3 ppbv in 1997/8 (Table 1). In contrast the measured annual mean SO_2 levels appear to have remained steady, or possibly even decreased slightly over the same period (Table 1), a period when large electricity generating plant in Malaysia has increasingly relied on natural gas (consumption up from 2.5 Mt in 1992 to 5.1 Mt in 1995) rather than fuel oil (consumption down from 2.4 Mt in 1992 to 2.0 Mt in 1995; DoE 1996b). This implies a stabilisation of sulfur emissions and increase in NO_x emissions from large stationary sources. Therefore, at least qualitatively, the time series data on the primary emissions (NO_x and SO_2) appear to reflect known changes in emissions strengths.

The rôle of base cation deposition in the atmospheric acid–base balance at PJ merits comment. In other parts of Asia, especially India and China, base cation deposition is very high, reducing considerably the net acid deposition (WMO, 1997; Larssen et al., 1998; Zhao et al., 1994). This is not the case at PJ, where the aerosol and rainwater data (Tables 2, 3) show base cation levels to be much less significant at this site. The low levels of dust-derived base cations almost certainly reflect the absence of deserts in the humid equatorial region in which PJ is located, suggesting a quantitative difference in the atmospheric acid–base mix in equatorial vs mid-latitude Asia.

A final comment concerns the potential environmental consequences of elevated levels of atmospheric acidity. The RAINS-Asia analysis in terms of critical loads is focussed on potential effects of terrestrial ecosystems, however a second area of potential effect is enhanced corrosion of building materials. According to Kucera and Fitz (1995), controlling variables determining rates of corrosion for a range of building materials include time of wetness, SO_2 mixing ratio and H^+ concentration in rainwater. Based on the observations presented above, the latter two variables are clearly enhanced at the PJ site in comparison with the unpolluted NT data. Moreover, based on the monthly mean meteorological data for PJ presented in the Klang Valley air quality assessment (JICA, 1993) the time of wetness parameter defined as the fraction of time with temperature >273K and relative humidity $>80\%$ would be non-zero frequently. The potential therefore is for significant corrosion rates under Klang Valley conditions. However we are unaware of any published work on corrosion for this region.

7. Conclusions

Time series data on gas, and aerosol concentration as well as rainwater composition have been measured over a five year period (March 1993–March 1998) at PJ in the industrialised Klang Valley of Malaysia. Total annual deposition fluxes of oxidised nitrogen plus sulfur species were estimated to average more than 300 meq m^{-2} yr^{-1} at PJ, a value comparable with fluxes reported from regions of Europe and North America in which acidic deposition has been reported to have adversely affected regional ecosystems. This total oxidised N plus S flux at PJ was also more than $15\times$ higher than the equivalent sum derived from gas and rainwater data collected at unpolluted tropical locations in Australia, confirming the dominance of anthropogenic sources of atmospheric N and S in the Klang Valley.

Target loads for acid deposition available at $1^\circ \times 1^\circ$ resolution from the RAINS-Asia model (Amann and Dhoondia, 1994) are an order of magnitude below the 300 meq m^{-2} yr^{-1} deposition fluxes determined from the measurements of oxidised N and S species at PJ. However the broad-scale target loads have not been validated directly in the Klang Valley region, so must be seen as quite uncertain at this stage. Nevertheless, the possibility of adverse environmental impacts resulting from the high levels of acid deposition determined must be acknowledged. Therefore, the extension of this work in two areas seems warranted: (1) verification of the RAINS-Asia target loads by direct measurement and analysis of soil and surface water properties in the Klang Valley region, and (2) determination of atmospheric acid components at additional sites within and outside the Klang Valley to determine the geographic extent of the phenomenon.

8. Acknowledgments and disclaimer

This work is a contribution to the Deposition of Biogeochemically Important Trace Species

activity of the International Global Atmospheric Chemistry Project, a component of the International Geosphere–Biosphere Programme. The study was funded by Tenaga Nasional Berhad, BHP Limited, CSIRO and the Malaysian Meteorological Service. However the views expressed are those of the authors and do not necessarily reflect the views of the supporting Institutions, which do not accept responsibility in respect of any information or advice given in relation to or as a consequence of anything contained herein. The authors express their gratitude to Malaysian Meteorological Service personnel who diligently maintained the measurement program over the 5-year study period.

REFERENCES

Amann, M. and Dhoondia, J. 1994. *RAINS-Asia user's manual*. World Bank, Washington DC.

Arndt, R. L. and Carmichael, G. R. 1995. Long-range transport and deposition of sulfur in Asia. *Water, Air and Soil Pollution* **85**, 2283–2288.

Ayers, G. P. 1991. Atmospheric ccidification in the Asian region. *Env. Monitoring Assessment* **19**, 225–250.

Ayers, G. P. and Gillett, R. W. 1988. *Acidification in Australia*. In: SCOPE 36, *Acidification in tropical countries* (eds. Rodhe, H. and Hererra. R.). SCOPE 36. J. Wiley and Sons, Chichester, England, pp. 347–402.

Ayers, G. P., Gillett, R. W., Selleck, P., Warne, J. O., Huysing, P. and Forgan, B. W. 1993. A pilot study on rain-water composition at Darwin Airport. *Aust. Meteorol. Magazine* **42**, 143–150.

Ayers, G. P. and Yeung, K. K. 1996. Acid deposition in Hong Kong. *Atmos. Environ.* **30**, 1581–1587.

Ayers, G. P., Keywood, M. D., Gillett, R. W., Manins, P. C., Malfroy, H. and Bardsley, T. 1998a. Validation of passive diffusion samplers for SO_2 and NO_2. *Atmos. Environ*., in press.

Ayers, G. P., Fukuzaki, N., Gillett, R. W., Selleck, P. W., Powell, J. C. and Hara, H. 1998b. Thymol as a biocide in Japanese rainwater. *J. Amos. Chem.* **30**, 301–310.

Baldocchi, D. D., Hicks, B. B. and Camara, P. 1987. A canopy stomatal resistance model for gaseous deposition to vegetated surfaces. *Atmospheric Environment* **21**, 91–101.

Barrett, K., Seland, X., Foss, A., Mylona, S., Sandnes, H., Styve, H. and Tarrason, L. 1995. EMEP/MSC-W Report 1/95. *European transboundary acidifying air pollution*. 10 years calculated fields and budgets to the end of the first sulphur protocol.

Carmichael, G. R. et al. 1995. Observed regional distribution of sulfur dioxide in Asia. *Water, Air and Soil Pollution* **85**, 2289–2294.

Chadwick, M. J. and Hutton, M. (eds.) 1991. *Acidic deposition in Europe*. Stockholm Environment Institute, Sweden, 376 pp.

DoE, 1996a. Department of Environment. *Malaysia: Malaysia environmental quality report*.

DoE, 1996b. Department of Environment. *Malaysia: environmental quality data 1992–1995*.

Downing, R. J., Hettelingh, J.-P. and Smet, P. A. M. 1991. *Calculation and mapping of critical loads for Europe*. RIVM report no. 259101003. Coordination Center for Effects, National Inst. Public Health and Env. Protection, Bilthoven, Netherlands.

EPA, 1994. *Quality assurance handbook for air pollution measurement systems,* vol. V. *Precipitation measurement systems*. United States Environmental Protection Agency, Office of Research and Development, Washington, EPA/600R-94/038e.

EPA, 1995. *Acid deposition standard feasibility study report to congress*. US Environmental Protection Agency. Office of Air and Radiation, Acid Rain Division.

Erisman, J. W., Versluis, A. H., Verplanke, T. A. J. W., De Haan, D., Anink, D., Van Elzakker, B. G., Mennen, M. G. and Van Aalst, R. M. 1993. Monitoring the dry deposition of SO_2 in the Netherlands: results for grassland and heather vegetation. *Atmospheric Environment* **27A**, 1153–1161.

Ferm, M. 1991. *A sensitive diffusional sampler*. Report L91–172, IVL, Box 47086, 402 58 Göteborg, Sweden.

Foell, W. K. and Green, C. W. 1991. Acid rain in Asia: an economic, energy and emissions overview. *Proc. 2nd Workshop on acid rain in Asia*. Asian Inst. Technol., Bangkok.

Galloway, J. N. 1988a. Effects of acid deposition on tropical aquatic ecosystems. In: *Acidification in tropical countries*. Rodhe, H. and Hererra, R. (eds). SCOPE 36. J. Wiley and Sons, Chichester, England, pp. 141–166.

Galloway, J. N. 1988b. Atmospheric acidification : projections for the future. *Ambio* **18**, 161–166.

Galloway, J. N. 1995. Acid deposition: perspectives in time and space. *Water, Air and Soil Pollution* **85**, 15–24.

Gillett, R. W., Ayers, G. P. and Noller, B. N. 1990. Rainwater acidity at Jabiru, Australia, in the wet season of 1983/84. *Sci. Tot. Environ.* **92**, 129–144.

Gillett, R. W. and Ayers, G. P. 1991. The use of thymol as a biocide in rainwater samples. *Atmos. Environ.* **25A**, 2677–2681.

Herlihy, L. J., Galloway, J. N. and Mills, A. L. 1987. Bacterial utilization of formic and acetic acids in rainwater. *Atmospheric Environment* **21**, 2397–2402.

Hettelingh, J.-P., Downing, R. J. and De Smet, P. A. M. 1991. *Mapping critical loads for Europe*. CCE Technical report no. 1. Coordination Center for Effects, National Inst. Public Health and Env. Protection, Bilthoven, Netherlands.

Hettelingh, J.-P., Sverdrup and Zhao, D. 1995. Deriving critical loads for Asia. *Water, Air and Soil Pollution* **85**, 2565–2570.

Hicks, B. B., Baldocchi, D. D., Meyers, T. P., Hosker, Jr., R. P. and Matt, D. R. 1987. A preliminary multiple resistance routine for deriving dry deposition velocities from measured quantities. *Water, Air, and Soil Pollution* **36**, 311–330.

Hicks, B. B. and Meyers, T. P. 1991. *Program DryDep2*. NOAA/ATDD. PO Box 2456, Oak Ridge, TN 37831-2456, USA.

Hicks, B. B., Hosker, R. P. Jr, Meyers, T. P. and Womack, J. D. 1991. Dry deposition inferential measurement techniques (I). Design and tests of a prototype meteorological and chemical system for determining dry deposition. *Atmos. Environ.* **25A**, 2345–2359.

Hornung, M., Sutton, M. A. and Wilson, R. B. (eds.) 1995. *Mapping and modelling of critical loads for nitrogen — a Workshop report*. Proc. Grange-Over-Sands Workshop, Inst. Of Terrestrial Ecology, UK, ISNN 1 870393 24 4.

JICA, 1993. *Air quality management study for the Kelang Valley Region*. Final Report vol. 2. Japan International Cooperation Agency, Tokyo, Japan.

Kato, N. and Akimoto, H. 1992. Anthropogenic emissions of SO_2 and NO_x in Asia: emission inventories. *Atmos. Environ.* **26a**, 2997–3017.

Kucera, V. and Fitz, S. 1995. Direct and indirect air pollution effects on materials including cultural monuments. *Water, Air and Soil Pollution* **85**, 153–165.

Kuylenstierna, J. C. I., Cambridge, H., Cinderby, S. and Chadwick, M. J. 1995. Terrestrial ecosystem sensitivity to acidic deposition in developing countries. *Water, Air and Soil Pollution* **85**, 2319–2324.

Larssen, T., Xiong, X., Vogt, R., Seip, H. H., Liao, B. and Zhao, D. 1998. Studies of soils, soil water and stream water at a small catchment near Guiyang, China. *Water, Air and Soil Pollution* **101**, 137–162.

Likens, G. E., Keene, W. C., Miller, J. M. and Galloway, J. N. 1987. Chemistry of precipitation from a remote, terrestrial site in Australia. *J. Geophys. Res.* **92**, 13,299–13,314.

Løkke, H., Bak, J., Falkengren-Grerup, U., Finlay, R. D., Ilvesniemi, H., Nygaard, P. H. and Starr, M. 1996. Critical loads of acidic deposition for forest soils is the current approach adequate? *Ambio* **25**, 510–516.

McDowell, W. H. 1988. Potential effects of acid deposition on tropical ecosystems. In: *Acidification in tropical countries*. Rodhe, H. and Hererra, R. (eds.). SCOPE 36. J. Wiley and Sons, Chichester, England, pp. 117–140.

Meyers, T. P., Hicks, B. B., Hosker, R. P., Womack, J. D. and Satterfield, L. C. 1991. Dry deposition inferential measurement techniques (II). Seasonal and annual deposition rates of sulfate and nitrate. *Atmos. Environ.* **25A**, 2361–2370.

MMS, 1988. *On rain acidity analysis based on data from the national acid rain monitoring network*. Malaysian Meteorological Service Report, Kuala Lumpur.

Mulder, J. and Stein, A. 1994. The solubility of aluminium in acid forest soils: long-term changes due to acid deposition. *Geochim. Cosmochim. Acta* **58**, 85–94.

Müller-Edzards, C., De Vries, W. and Erisman, J. W. (eds.) 1997. *Ten years of monitoring forest condition Europe: studies on temporal development, spatial distribution and impacts of natural and anthropogenic stress factors*. EC and UN/ECE Technical Report, Brussels, Geneva.

NAPAP, 1991. *Acid deposition: state of science and technology*. National Acid Precipitation Assessment Program Office of the Director, Washington, DC, p. 2053.

Nilsson, J. and Grennfelt, P. (eds.) 1988. *Critical loads for sulfur and nitrogen*. Miljörapport 1988: 15, Nordic Council of Ministers.

Noller, B. N., Currey, N. A., Ayers G. P. and Gillett, R. W. 1990. Chemical composition and acidity of rainfall in the Alligator Rivers region, Northern territory, Australia. *Sci. Tot. Environ.* **91**, 23–48.

Overrein, L. O., Seip, H. M. and Tollan, A. 1980. *Acid precipitation — effects on forest and fish*. Final report from SNSF-project 1972–1980, FR 19/80. Norwegian Institute for Water Research, Oslo.

RMCC, 1992. Federal/Provincial Research and Monitoring Committee, *The 1990 Canadian long-range transport of air pollutants and acid deposition assessment report*. Environment Canada, Toronto, Canada.

Rodhe, H. and Hererra, R. (eds.) 1988. *Acidification in tropical countries*. SCOPE 36. J. Wiley and Sons, Chichester, England.

Rodhe, H., Galloway, J. N. and Zhao, D. 1992. Acidification in Southeast Asia — prospects for the coming decades. *Ambio* **21**, 148–150.

Rodhe, H. et al. 1995a. Acid rain '95? Summary Statement. *Water, Air and Soil Pollution* **85**, 1–14.

Rodhe, H., Langner, J., Gallardo, L. and Kjellström, E, 1995b. Global scale transport of acidifying pollutants. *Water, Air and Soil Pollution* **85**, 37–50.

Seip, H. M., Anderson, D. O., Christopherson, N., Sullivan, T. J. and Vogt, R. D. 1989. Variations in concentrations of aqueous aluminium and other chemical species during hydrological episodes at Birkenes southernmost Norway. *J. Hydrol.* **108**, 387–405.

Schlünzen, K. H. and Pahl, S. 1992. Modification of dry deposition in a developing sea-breeze circulation — a numerical case study. *Atmospheric Environment* **26A**, 51–61.

Stull, R. B. 1988. *An introduction to boundary layer meteorology*. Kluwer Academic Publishers, Dordrecht, The Netherlands.

Walcek, C. J., Brost, R. A. and Chang, J. S. 1986. SO_2, sulfate and HNO_3 deposition velocities computed using regional land use and meteorological data. *Atmospheric Environment* **20**, 949–964.

Wesely, M. L., Cook, D. R. and Hart R. L. 1983. Fluxes of gases and particles above a deciduous forest in wintertime. *Boundary-Layer Meteorology* **27**, 237–255.

WMO, 1992. *Report of the WMO meeting of experts on the quality assurance plan for the global atmosphere*

watch. Garmisch-Partenkirchen, Germany, 26–30 March, 1992, WMO/GAW no. 80.

WMO, 1997. *Global acid deposition assessment*. World Meteorological Organization report TD no. 777, Whelpdale, D. M., Kaiser M. S. (eds.).

Zhao, D. and Seip, H. M. 1991. Assessing the effects of acid deposition in southwestern China using the MAGIC model. *Water Air and Soil Pollution* **60**, 83–97.

Zhao, Dawei, Seip, H. M., Zhao Dianwu and Zhang, D. 1994. Pattern and cause of acidic deposition in Chongqing region, Sichuan Province, China. *Water Air and Soil Pollution* **74**, 1–22.