

# Indoor Exposures to Acidic Aerosols at Child and Elderly Care Facilities

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## Abstract

*A six-week study of indoor and outdoor air pollutants was conducted in central New Jersey during the summer months of 1989. Three institutional settings for elderly and child care were investigated for the potential of acidic aerosol exposures. The indoor penetration by fine aerosols was > 70% at all three institutions. For locations with closed ventilation, it was 15-25% lower than for the open-window setting. Relative to outdoor levels, indoor acidic sulfate aerosols were 30-57% neutralized. Indoor levels of ammonia were  $\approx 10 \times$  higher than corresponding outdoor values, which were consistent with calculated emission rates from human occupants. From estimates of total daily exposure, 75% of the daily dose of aerosol acidity for the elderly was due to indoor exposures. Doses received by the elderly and children ranged from 290 to 1100 nmoles of acid (15 to 55  $\mu\text{g}$  as  $\text{H}_2\text{SO}_4$ ) in a 24-h period with "worst-case" dose received by children as high as 3400 nmoles of acid in the daytime. These doses were comparable to the levels observed in clinical and epidemiological studies where health effects result. The daily dose of acid delivered to children was calculated to be 2 to 4  $\times$  higher than the dose to the elderly population. The calculations for children indicate that more than 90% of their dose on a summer day may come from outdoor exposures. These data suggest that indoor settings are protective, but children may still be at risk from summertime acidic aerosol exposure, depending on their activities outdoors.*

## KEY WORDS:

Indoor air, Acidic aerosol, Ammonia neutralization, Human exposure, Institutions.

Manuscript received: 3 October 1991

Accepted for publication: 17 October 1992

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## Introduction

A summertime study of acidic aerosol exposures was conducted at three different institutional settings in New Jersey during the summer of 1989. Indoor and outdoor concentrations and exposure relationships were investigated by a concurrent sampling program lasting six weeks. The objectives of this study were to understand (1) the temporal pattern of indoor acidic aerosol concentrations in institutional settings and (2) the potential exposures to sensitive populations, such as the elderly and children.

The assessment of acidic aerosol exposure has increased in recent years due to growing evidence, direct and indirect, of their adverse health effects observed in clinical (Koenig et al., 1989; Schlesinger, 1989; USEPA, 1989a) and epidemiological investigations (Bates and Sizto, 1989; Ostro et al., 1991; Ware et al., 1986). Outdoor concentrations in the range of 20-200 nmole/m<sup>3</sup> (1-10  $\mu\text{g}/\text{m}^3$  as  $\text{H}_2\text{SO}_4$ ) for a 24-hour period are commonly monitored in various locations in the northeast of U.S. and Canada (Lioy and Waldman, 1989). Summertime levels are generally higher than those measured during the other months (Tanner et al., 1981; Waldman et al., 1990; Keeler et al., 1990; Pierson et al., 1989).

Since the 1970's, research has focused on indoor-outdoor relationships of pollutants. The results show that indoor exposures to air pollutants may differ substantially from those outdoors (Yocum, 1982). Different patterns for indoor air quality occur for species with, versus those without, indoor sources.

In a recent study, Brauer et al. (1991) monitored acidic aerosols and gases in 11 houses in the Boston area during summer and winter 1989. Daily 24-h samples were collected concurrently at indoor and outdoor locations during the study periods. Indoor levels of acidic aerosols in homes had a range 0-40 nmole/m<sup>3</sup> (0-2  $\mu\text{g}/\text{m}^3$  as  $\text{H}_2\text{SO}_4$ ). Infiltration of sulfate aerosol averaged 0.96 of outdoor levels, and 40-50% of outdoor aerosol acidity was neutralized dur-

ing infiltration. None of the residences were air-conditioned. High levels of indoor ammonia were also observed. Nevertheless, acidic aerosol was found to coexist with indoor ammonia. Another study by Li and Harrison (1990) examined office buildings with air-conditioning during the summer. The infiltration to office buildings was found to be slightly lower than to the Boston houses; indoor/outdoor ratios of  $\text{SO}_4^-$  were 0.82. Factors such as ventilation system and building type are important in determining indoor levels of acidic aerosol exposure.

## Method

The Indoor Denuder Sampler (IDS) used in this study is a modified version of the Annular Denuder System (ADS) (Possanzini et al., 1983; Brauer et al., 1989). The IDS utilizes the ADS inlet/impactor with a  $2.5 \mu\text{m}$  particle cut-size (to remove coarse particles), a single glass annular denuder tube, followed by a dual-filter pack. The IDS denuders are coated with citric acid to collect gaseous ammonia and prevent aerosol neutralization on the filter substrate. Aerosols are collected on Teflon membrane filters

(Gelman Teflo,  $1 \mu\text{m}$  pore size). Backup Nylon filters (Gelman Nylosorb) trap nitric acid in the sampling stream. Hence, the IDS gives measurements of aerosol species, gaseous ammonia and nitric acid. The air sampling pump is configured with a solenoid valve and timer which allows two 12-h integrated samples per day (day and night) with a single daily setup visit to the study site. The connector fitted to the base of the filter holder enables quick replacement of the IDS at the field locations.

The sampling strategy was designed to determine acid aerosol exposures encountered by the elderly and pre-school children at institutional settings. A day-care center (DC), nursing home (LN) and home for elderly (MH) were located short distances from a central outdoor monitoring site (RC) (see Figure 1). The 2-story building at site DC provided day-care facilities on the first floor for about 50 children. The sampler was placed in a recreation room ( $60 \text{ m}^3$ ) leading to the main entrance of the first floor; windows were kept closed and a window air-conditioner operated constantly with closed ventilation. The LN site was a modern, single-story building with central air-conditioning and housed 100 elderly patients.

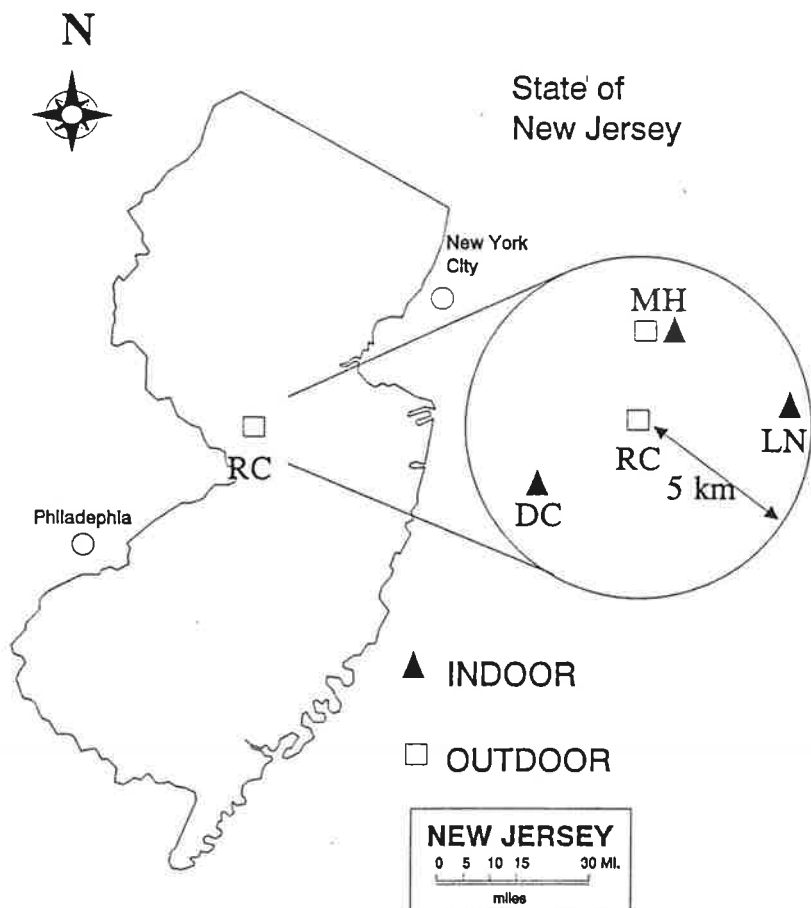


Fig. 1 Location of sampling sites in acidic aerosol exposure study (see text for descriptions of sites).

**Table 1** Minimum Quantitative Limits (MQL) for component measurements

Species	MQL (neq/m <sup>3</sup> )	Precision uncertainty
<b>Aerosol</b>		
H <sup>+</sup>	7 (0.4 µg/m <sup>3</sup> as H <sub>2</sub> SO <sub>4</sub> )	7%
NH <sub>4</sub> <sup>+</sup>	7	13%
SO <sub>4</sub> <sup>=</sup>	30 (1.5 µg/m <sup>3</sup> )	2%
NO <sub>3</sub> <sup>-</sup>	15	15%
NO <sub>2</sub>	7	15%
<b>Gaseous</b>		
NH <sub>3</sub>	7 (0.2 ppb)	13%
SO <sub>2</sub>	30 (0.9 ppb)	2%
HNO <sub>3</sub>	15 (0.5 ppb)	15%
HNO <sub>2</sub>	7 (0.2 ppb)	15%

$$MQL = \frac{C \times E}{V}$$

C: Lowest concentration of standards in analytical procedure

E: Volume of extraction

V: Volume of air sampled

The IDS was placed in a communal recreation room (92 m<sup>3</sup>). The third site (MH/MO) was an 80-year old two-story building with 45 elderly residents. There were no air-conditioners, and windows were kept open almost continuously during the study period. One IDS unit (MH) was placed indoors in the institutional dining room (350 m<sup>3</sup>). A second IDS (MO) measured outdoor concentrations and was situated on the second floor balcony immediately outside the dining room. This unit was 3 m from the outside wall of the building and 5 m above ground level. An ADS sampler was used at the central outdoor monitoring station (RC), located in an athletic field of a community college. This sampler was set up on the roof of a trailer about 4 m above ground level. The unit employed two types of denuders: carbonate-coated (to collect SO<sub>2</sub>, HNO<sub>3</sub> and HONO) plus citric acid-coated for ammonia. Outdoor ozone and local meteorological data (temperature, relative humidity, wind speed and wind direction) were also recorded at the RC site.

Daily monitoring took place between June 20 and July 30, 1989. The schedule for DC and LN was one 12-h sample each day (7:00 am to 7:00 pm local time), whereas two 12-h samples (7:00 am to 7:00 pm and 7:00 pm to 7:00 am local time) were taken at MH, MO and RC. Weekday samples only were collected at DC because the facility was closed at weekends. After every run, the ADS and IDS units were capped and replaced with fresh units. On return to the laboratory, denuder tubes and filters were immediately extracted with 10 ml solution. Denuder tubes were extracted with DI-H<sub>2</sub>O, and filters were handled exclusively in a NH<sub>3</sub>-free glove box.

Nylon filters were extracted in Borate/Gluconate buffer solution, which is identical to IC eluent for anion analysis. For the Teflon filters, the PTFE membrane was first cut away from the plastic support ring, then wetted with 10 µl methanol and extracted in HClO<sub>4</sub> solution (5 × 10<sup>-5</sup> M) for 30 minutes in an ultrasonic bath. The analyses for acidity were conducted with a micro-combination pH electrode and titrimeter; sulfuric acid standards were prepared in the HClO<sub>4</sub> solution. Ammonium was measured using the automated colorimetric indophenol method, and ionchromatography (IC) was used for anion determination.

Minimum Quantitative Limits (MQL) were based on the working sensitivity of the analytical instruments (5-20 µeq/l), the extraction volume (10 ml), and the volume of air sampled (≈ 7 m<sup>3</sup>). The corresponding MQL for the concentrations in air are listed in Table 1.

## Results

The aerosol and gaseous components measured from samples collected during the study period are presented in Table 2. Outdoor concentrations of acidic aerosol components were comparable to previous studies, while indoor levels were generally lower than outdoor values, except for ammonia. The indoor-outdoor ratios for aerosol and gaseous species are presented in Table 3. Ratios below one represent species with minimal or no indoor source (e.g. SO<sub>2</sub>, SO<sub>4</sub><sup>=</sup>). In this case, the ratio characterizes the penetration of outdoor materials to the indoor microenvironments. In cases of reactive compounds (e.g. HNO<sub>3</sub>), it represents the lower limit of penetration. On the other hand, larger (than one) ratios show the presence of a dominating indoor source for such species.

### Sulfate and Sulfur Dioxide

Outdoor levels of sulfate ranged between 30 to 490 neq/m<sup>3</sup> with mean concentrations of 197 (MO) and 154 (RC). The difference in means was due to the larger data set at RC. The difference between the two outdoor locations was insignificant when compared with paired t-test at the level p < 0.01. The levels of sulfate aerosol at two outdoor sites were well correlated for both daytime and night-time samples (Table 4). Furthermore, outdoor air pollution was clearly discernible in the records of indoor sulfate levels (Figure 2). Daytime sulfate levels were on average 15% higher than night-time levels (164 vs 142 nmole/m<sup>3</sup> at RC and 205 vs 188 nmole/m<sup>3</sup> at MO).

**Table 2** Acidic aerosols and gases at outdoor (RC and MO) and indoor (MH, LN & DC) sites. Aerosols in neq/m<sup>3</sup> and gases in ppb.

	M ± S.E.	GM	90%-10%		M ± S.E.	GM	90%-10%
<b>RC (n = 29/31)</b>				<b>MH (n = 41/28)</b>			
H <sup>+</sup>	51 ± 7	30	122-<MQL	H <sup>+</sup>	23 ± 3	15	54-<MQL
(D)	66 ± 10	34	135- 12	(D)	21 ± 4	12	53-<MQL
(N)	31 ± 7	25	71-<MQL	(N)	29 ± 5	24	66-<MQL
SO <sub>4</sub> <sup>-</sup>	154 ± 20	98	341-<MQL	SO <sub>4</sub> <sup>-</sup>	171 ± 20	108	398- 36
(D)	164 ± 20	102	341-<MQL	(D)	173 ± 22	111	407- 34
(N)	142 ± 20	84	300-<MQL	(N)	163 ± 29	96	398- 38
NH <sub>4</sub> <sup>+</sup>	131 ± 10	98	292- 27	NH <sub>4</sub> <sup>+</sup>	115 ± 10	77	282-<MQL
(D)	136 ± 20	111	258- 27	(D)	117 ± 17	89	299-<MQL
(N)	128 ± 20	88	292- 25	(N)	108 ± 19	64	270- 8
NO <sub>3</sub> <sup>-</sup>	15 ± 2	12	33-<MQL	NO <sub>3</sub> <sup>-</sup>	0.9 ± 0.3	0.0	4-<MQL
(D)	13 ± 2	11	33-<MQL	(D)	0.5 ± 0.2	0.0	2-<MQL
(N)	17 ± 3	14	32-<MQL	(N)	1.6 ± 0.7	0.0	6-<MQL
H <sup>+</sup> /SO <sub>4</sub> <sup>-</sup>	0.40 ± 0.04	0.41	0.73-0.04	H <sup>+</sup> /SO <sub>4</sub> <sup>-</sup>	0.18 ± 0.03	0.14	0.47-<MQL
(D)	0.47 ± 0.05	0.43	0.75-0.10	(D)	0.17 ± 0.04	0.06	0.48-<MQL
(N)	0.34 ± 0.04	0.37	0.58-0.04	(N)	0.20 ± 0.03	0.17	0.57-<MQL
NH <sub>3</sub>	3.3 ± 0.4	2.6	5.2-1.4	NH <sub>3</sub>	29 ± 4	19	42- 7
(D)	3.1 ± 0.3	2.5	4.9-1.1	(D)	31 ± 6	19	42- 9
(N)	3.5 ± 0.6	2.6	5.2-1.4	(N)	29 ± 5	21	60-14
HNO <sub>3</sub>	1.6 ± 0.2	0.8	4.6-<MQL	HNO <sub>3</sub>	0.4 ± 0.05	0.3	0.9-<MQL
(D)	2.5 ± 0.3	1.8	5.1-<MQL	(D)	0.4 ± 0.08	0.3	1-<MQL
(N)	0.6 ± 0.1	0.4	1.4-<MQL	(N)	0.2 ± 0.05	0.2	1-<MQL
SO <sub>2</sub>	6.0 ± 0.6	3.7	14-1.3	Temp. (°C)	26.7 ± 1	26.1	31.1-21.1
(D)	6.9 ± 0.9	4.7	15-1.3				
(N)	4.9 ± 0.9	2.9	12-1.2				
HNO <sub>2</sub>	1.2 ± 0.09	1.1	2.2-0.2	<b>LN (n = 37/0)</b>			
(D)	0.8 ± 0.09	0.7	1.8-<MQL	H <sup>+</sup>	17 ± 3	10	42-<MQL
(N)	1.7 ± 0.10	1.7	2.7-0.5	SO <sub>4</sub> <sup>-</sup>	165 ± 21	104	380- 28
				NH <sub>4</sub> <sup>+</sup>	73 ± 13	49	197-<MQL
				NO <sub>3</sub> <sup>-</sup>	13 ± 1	11	22- 7
				H <sup>+</sup> /SO <sub>4</sub> <sup>-</sup>	0.15 ± 0.03	0.09	0.39-<MQL
				NH <sub>3</sub>	56 ± 5	59	85-15
				HNO <sub>3</sub>	0.3 ± 0.1	0.2	1-<MQL
				Temp (°C)	25.0 ± 1	25.0	27.2-22.8
Daily 1-h peak				<b>DC (n = 24/0)</b>			
O <sub>3</sub>	68 ± 3	62	102-35	H <sup>+</sup>	15 ± 4	11	44-<MQL
Temp. (°C)	22.8 ± 2	22.2	27.8-17.2	SO <sub>4</sub> <sup>-</sup>	173 ± 30	135	342- 32
(D)	26.1 ± 3	26.1	32.8-17.8	NH <sub>4</sub> <sup>+</sup>	92 ± 19	71	234- 8
(N)	19.4 ± 3	20.0	26.1-12.8	NO <sub>3</sub> <sup>-</sup>	12 ± 2	11	25-<MQL
				H <sup>+</sup> /SO <sub>4</sub> <sup>-</sup>	0.16 ± 0.05	0.06	0.45-<MQL
				NH <sub>3</sub>	61 ± 9	65	114- 6
				HNO <sub>3</sub>	0.3 ± 0.07	0.0	1-<MQL
				Temp (°C)	26.1 ± 1	26.1	28.9-22.2
<b>MO (n = 39/32)</b>				D: daytime sample; N: night-time sample; n: number of D/N samples; m ± S.E.: mean ± standard error; GM: geometric mean; 90%-10%: range; MQL: minimum quantitative level			
H <sup>+</sup>	47 ± 7	32	103-<MQL	<b>Outdoor sulfur dioxide measured at RC ranged between 1 to 18 ppb with a mean 12-h period value of 6 ± 0.8 ppb. As is the case with sulfate, the higher levels occurred in the daytime; SO<sub>2</sub> was strongly correlated with sulfate (p &lt; 0.0001).</b>			
(D)	54 ± 12	32	152-<MQL	<b>The indoor/outdoor (I/O) ratios of sulfate aerosol were close to one, 0.92 at MH and decreased to 0.76 and 0.67 at DC and LN, respectively. One reason for the higher I/O ratios at MH is that open windows and doors were used for ventilation versus air-conditioning (closed ventilation) systems in the other</b>			
(N)	39 ± 8	33	94-<MQL				
SO <sub>4</sub> <sup>-</sup>	197 ± 20	114	497- 34				
(D)	205 ± 28	138	483- 34				
(N)	188 ± 32	111	497- 41				
NH <sub>4</sub> <sup>+</sup>	131 ± 20	97	327-<MQL				
(D)	148 ± 28	113	327-<MQL				
(N)	113 ± 21	82	327-<MQL				
NO <sub>3</sub> <sup>-</sup>	6 ± 2	0	11-<MQL				
(D)	4 ± 3	0	7-<MQL				
(N)	9 ± 4	0	22-<MQL				
H <sup>+</sup> /SO <sub>4</sub> <sup>-</sup>	0.49 ± 0.11	0.17	0.88-<MQL				
(D)	0.47 ± 0.14	0.20	0.96-<MQL				
(N)	0.51 ± 0.19	0.14	0.86-<MQL				
NH <sub>3</sub>	9 ± 1	6	23- 3				
(D)	10 ± 2	6	24- 3				
(N)	9 ± 1	6	21- 3				
HNO <sub>3</sub>	1.1 ± 0.2	0.8	2.4-0.2				
(D)	1.4 ± 0.2	1.1	3-0.4				
(N)	0.8 ± 0.3	0.5	1-0.2				
Temp.: same as RC outdoor site							

... continued

**Table 3** Indoor-to-outdoor concentration ratios

		I/O Ratio	
		GM	90%-10%
<b>MH/MO</b>			
H <sup>+</sup>	(D)	0.47	1.00-0.00
	(N)	0.73	1.08-0.23
SO <sub>4</sub> <sup>-</sup>	(D)	0.91	1.16-0.56
	(N)	0.93	1.31-0.59
NH <sub>3</sub>	(D)	4.84	13.30-0.42
	(N)	4.22	8.08-0.84
NH <sub>4</sub> <sup>+</sup>	(D)	1.08	2.16-0.00
	(N)	1.00	2.16-0.31
NO <sub>3</sub>	(D)	0.11	0.43-0.00
	(N)	0.44	0.50-0.00
HNO <sub>3</sub>	(D)	0.29	0.67-0.00
	(N)	0.37	1.00-0.00
<b>LN/MO</b>			
H <sup>+</sup>	(D)	0.40	0.96-0.00
SO <sub>4</sub> <sup>-</sup>	(D)	0.74	1.16-0.52
NH <sub>3</sub>	(D)	10.60	21.20-1.15
NH <sub>4</sub> <sup>+</sup>	(D)	0.44	0.89-0.00
NO <sub>3</sub>	(D)	1.37	2.00-0.12
HNO <sub>3</sub>	(D)	0.12	0.28-0.00
<b>DC/MO</b>			
H <sup>+</sup>	(D)	0.46	1.33-0.00
SO <sub>4</sub> <sup>-</sup>	(D)	0.82	1.07-0.57
NH <sub>3</sub>	(D)	9.80	20.70-1.63
NH <sub>4</sub> <sup>+</sup>	(D)	0.80	1.98-0.00
NO <sub>3</sub>	(D)	1.78	4.75-0.00
HNO <sub>3</sub>	(D)	0.09	0.32-0.00

D: daytime sample; GM: geometric mean  
N: night-time sample; 90%-10%: range

two buildings. At the day-care center (DC), the sampler was located in an area near the entry, and at LN, it was away from the entry and in an area with less human traffic, which is consistent with the lowest indoor/outdoor ratio observed there.

### Aerosol Acidity and Ozone

For the outdoor sites, the mean daytime acidity was 60 neq/m<sup>3</sup> (3 µg/m<sup>3</sup> as H<sub>2</sub>SO<sub>4</sub>) and the night-time acidity was 35 neq/m<sup>3</sup>. The peak outdoor concentration was 400 neq/m<sup>3</sup> for a 12-h period. There were

eight periods during the study when 12-h average values were >100 neq/m<sup>3</sup> and the elevated concentrations lasted up to 36-h. Similar to sulfate aerosol, there was a diurnal pattern for aerosol acidity, with values ≈ 60% higher in the daytime compared to the following night-time periods.

The mean outdoor H<sup>+</sup>/SO<sub>4</sub><sup>-</sup> equivalent ratio was 0.4, indicating a mixture of ammonium, sulfate and strong acidity in the form slightly more neutralized than ammonium bisulfate [(NH<sub>4</sub>)HSO<sub>4</sub>]. Concentrations were significantly correlated between the two outdoor locations (Table 4). Paired t-test showed an insignificant difference between outdoor sites at the level of p < 0.01, indicating that acidic sulfate aerosol was homogeneously distributed over a scale of ≈ 5 km.

Mean indoor acidity was less than 20 neq/m<sup>3</sup> for all indoor locations, with the indoor 12-h peak being 60 neq/m<sup>3</sup>. Among the indoor locations, aerosol acidity for the air-conditioned sites (DC and LN) were lower than at the open-window location (MH). In contrast to the outdoor observations, indoor aerosol acidity at MH was higher during the night-time.

The ratios of indoor/outdoor acidity were much lower than the corresponding sulfate values. The mean I/O ratios at DC and LN were 0.46 and 0.40 respectively, while at MH the I/O ratios were 0.47 and 0.73, for daytime and night-time periods. The acidity measurements for daytime MH and DC samples showed no correlation with the corresponding outdoor acidity. For night-time MH and LN samples, the correlations between indoor and outdoor levels were highly significant (p < 0.0001). The night-time MH/LN datasets represent the periods or locations for which little human activity occurred near the monitor.

The indoor sites were devoid of fine aerosol sources; thus any aerosol acidity monitored indoors was due to the infiltration of outdoor acidic sulfate particles. Hence, we can express the neutralization of the

**Table 4** Correlation coefficients of concentrations among sampling sites

Sites	Sulfate	Acidity	Ammonium	Ammonia	Nitrate	Nitric Acid
RC/MO	***	***	***	ns	ns	**
(D)	***	**	**	ns	ns	***
(N)	***	*	*	ns	ns	ns
DC/MO (D)	*	ns	**	ns	ns	ns
LN/MO (D)	**	***	***	ns	ns	ns
MH/MO	***	**	***	ns	ns	**
(D)	***	ns	***	ns	ns	*
(N)	***	***	***	ns	ns	ns

\*\*\*: p < 0.0001 \*\*; p < 0.001 \*: p < 0.01 ns: non-significant, p > 0.01

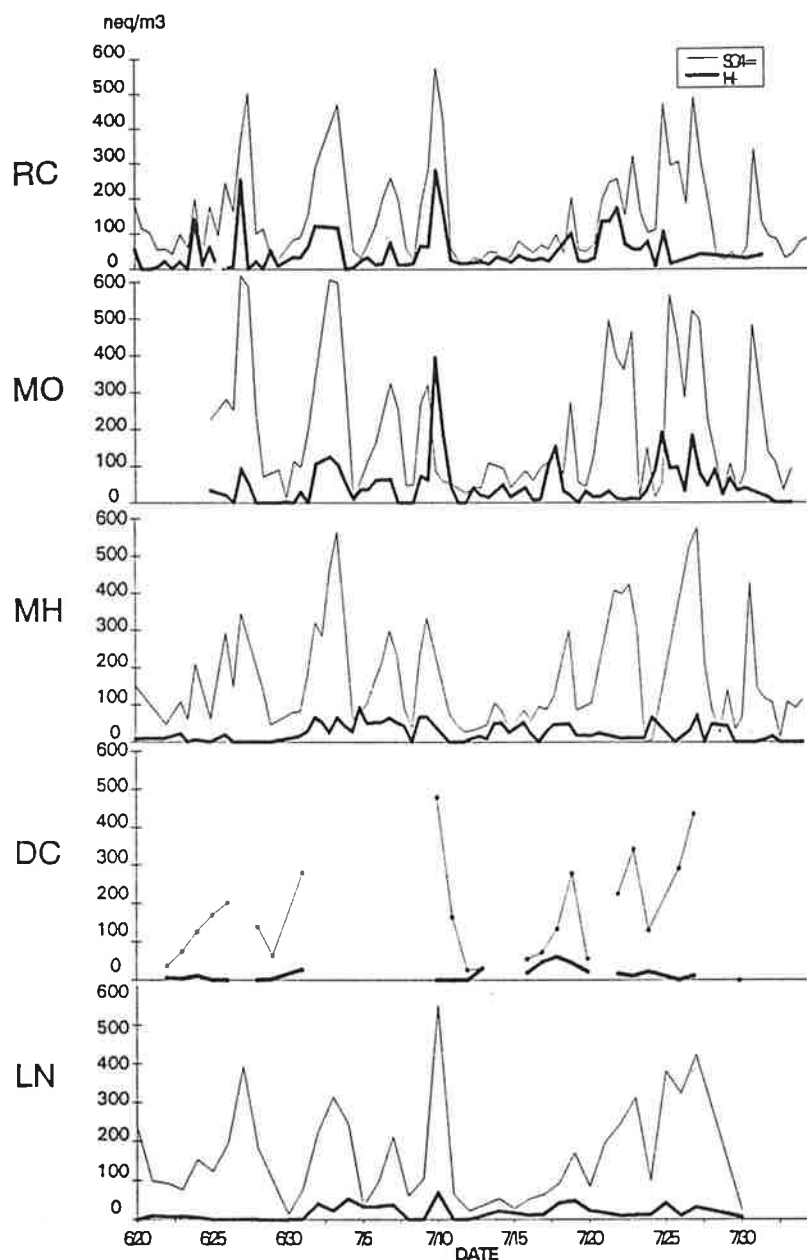


Fig. 2 Time series of sulfate and acidity for RC (central outdoor), MO (local outdoor) and three indoor locations (MH, DC and LN).

acidic sulfate aerosols in terms of the I/O ratios of acidity and sulfate, as follows:

$$\% \text{ neutralization} = \left(1 - \frac{I/O (H^+)}{I/O (SO_4^-)}\right) \times 100 \quad (1)$$

The neutralization at the indoor locations ranged between 30% and 57% (Table 5). However, large standard deviations indicate that the neutralization may vary greatly during a 12-h sampling period. A lower degree of neutralization was observed in the night-time periods at MH, while daytime periods were similar for all three indoor locations.

Ozone levels presented in Table 2 are 1-h peak concentrations observed at RC site. Daily peak values ranged between 9 to 150 ppb with a mean value during the study period of 68 ppb. Ozone concentrations were correlated with outdoor acidity but not with indoor acidity at any of the three indoor locations. Other factors such as temperature,  $HNO_3$  and  $SO_4^-$  were also found to be strongly correlated with ozone levels ( $p < 0.0001$ ).

#### Ammonia and Ammonium

Outdoor ammonia ( $NH_3$ ) concentrations at the central outdoor (RC) site ranged from 2 to 6 ppb with

**Table 5** Percentage of neutralization for indoor aerosol acidity samples

Site	Time	Median	Mean $\pm$ std.
MH	Day	59	52 $\pm$ 46
	Night	28	30 $\pm$ 42
LN	Day	62	42 $\pm$ 62
DC	Night	68	57 $\pm$ 54

night-time ammonia slightly higher than daytime levels. The indoor concentrations were much larger, with median values ranging between 20 and 60 ppb and peak 12-h concentrations as high as 170 ppb. In contrast to the outdoor levels, higher concentrations during the daytime were observed. Ammonia levels were significantly higher at DC and LN compared to MH (Figure 3).

The acidic fraction of aerosols ( $H^+/SO_4^{2-}$ ) among sites was found to be inversely related to ammonia levels (Figure 3). The capacity for these levels of indoor ammonia to neutralize acidic aerosol was in the range of 400-1200 neq/m<sup>3</sup> (20-60  $\mu$ g/m<sup>3</sup> as H<sub>2</sub>SO<sub>4</sub>), far higher than peak concentrations outdoors. As shown in Figure 3, the presence of such high ammonia concentrations reduced the acid fraction of sulfate, although the acidity was not eliminated entirely.

### Nitric Acid and Nitrate

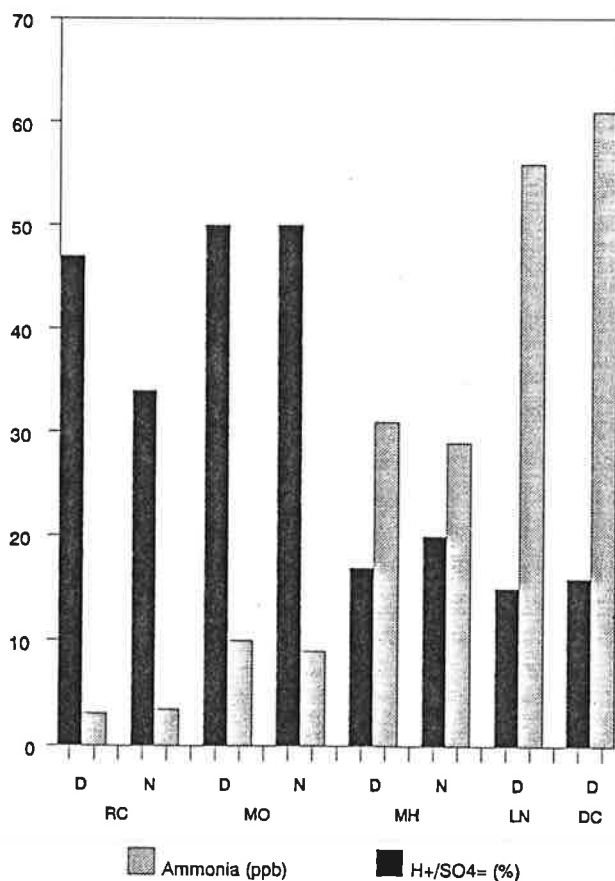
Outdoor nitric acid concentrations averaged 1.6 and 1.1 ppb at RC and MO, respectively. Nitric acid was strongly correlated with the levels of acidic sulfate aerosol ( $p < 0.001$ ), indicating that both species are associated with the same source or with local meteorology. Lower levels of nitric acid were measured at MO than at RC. This could be due to the rapid deposition of nitric acid to surfaces, which may have occurred since the MO sampler was very close to the building.

Indoor levels of nitric acid were much lower than outdoor levels with the mean indoor nitric acid level being below the MQL (0.5 ppb) at all sites. The I/O ratios for nitric acid were  $< 0.3$  at indoor locations (Table 3), indicating that there was a substantial fraction lost during penetration into indoors. As expected, the I/O ratios were lowest for DC and LN sites. At both outdoor and indoor sites, clear diurnal patterns of nitric acid were observed. Daytime levels were 60% higher than night-time values.

Low concentrations of the nitrate aerosol were measured at all sites with the mean outdoor concentrations being 12 and 6 neq/m<sup>3</sup> for RC and MO sites, respectively. The indoor concentrations were lower

than outdoor levels, with mean values for indoor locations below the MQL. Due to substantial loss of nitric acid during penetration and low indoor nitrate levels (smaller than MQL), the indoor ammonium nitrate system ( $NH_4NO_3(s) \rightleftharpoons HNO_3(g) + NH_3(g)$ ) could not achieve equilibrium in half of the sampling periods. The agreement between theoretical and measured values of the ammonia-nitric acid partial pressure product was poor. Based upon the nitrate data higher than MQL, we found that indoor concentrations at the MH site were significantly lower than those at the LN and DC sites (paired t-test  $p < 0.01$ ). The I/O ratios for the aforementioned data set were 0.11 and 0.44 for MH (day and night), while for LN and DC they were 1.4 and 1.8, respectively.

At the outdoor sites and MH, the fraction of aerosol nitrate ( $NO_3^-/N_T$ ) was generally low. Furthermore, lower fractions were observed for the daytime periods than for night-time periods, with the day-



**Fig. 3** Average concentrations of ammonia (ppb) and acid-sulfate fraction (as percentage) for sites of 12-h samples. RC and MO were outdoor sites, MH, LN and DC were indoor sites. D and N represent day (7:00 am to 7:00 pm) and night (7:00 pm to 7:00 am) samples.



**Table 6** Fraction of aerosol nitrate to total nitrate for different sampling locations

Site	RC	MO	MH	LN	DC
NO <sub>3</sub> /N <sub>T</sub>					
(D)	0.13	0.05	0.02	0.66	0.61
(N)	0.42	0.17	0.12	---	---

time temperature on average being 7 °C higher than the night-time temperature. The mean values (NO<sub>3</sub>/N<sub>T</sub>) for MH and MO were similar, while the mean values at RC were slightly higher than at MO (Table 6). At LN and DC sites, over than 50% of N<sub>T</sub> was in the aerosol form. These two indoor environments had air-conditioning which apparently produced conditions more favorable for nitrate aerosol formation, interior temperature being an important factor.

## Discussion

Two temporal patterns were observed for aerosol acidity: for outdoors, higher levels of both sulfate and acidity were observed in the daytime; for indoors, while sulfate was higher during the daytime, higher acidity levels were observed in the night-time. Higher daytime sulfate levels are chiefly due to photochemical reactions affecting SO<sub>2</sub> oxidation (USEPA, 1982). For indoor environments, however, the diurnal pattern may have been the outcome of penetration of outdoor acidic aerosols, with neutralization by indoor ammonia. The penetration rates were about the same, day and night, while the neutralization indoors was lower for the night-time periods.)

The variation of the acidity concentrations measured in LN and DC versus MH demonstrates the influence of the ventilation system on indoor levels. Several factors contribute to this. First, closed-circulation ventilation systems reduce the effective penetration of outdoor particles and gases. The air-conditioners capture and remove sulfate aerosol directly, and they also decrease the air exchange rate by recirculating the air. Second, the closed-circulation system promotes acidic aerosol neutralization: it leads to higher levels of ammonia and increases the residence time for the neutralization reaction to proceed.

## Penetration

In the present study, penetration of outdoor fine aerosol to indoor environments (based on sulfate aerosol measurements) is qualitatively demonstrated by I/O ratios of 0.92, 0.67 and 0.76 for MH, LN and

DC, respectively. The penetration efficiency at MH was virtually the same for the daytime and night-time periods. Air-conditioned buildings (LN & DC) showed 15-25% less efficient penetration than the open-window setting (MH). The high indoor/outdoor ratios in all three cases indicate that infiltration of outdoor air was highly effective and had the dominant effect on the accumulation of aerosol acidity indoors.

The I/O ratios for institutional settings are similar to those found in recent studies by Brauer et al. (1991) for residential buildings (I/O = 0.96) and by Li and Harrison (1990) for university buildings (I/O = 0.81). A study by Moschandreas et al. (1981) found an average sulfate I/O of 0.70 for both office buildings and residences without gas stoves. In Harvard's Six Cities Study, the use of air-conditioners reduced I/O ratios of sulfates by approximately 50% (Dockery and Spengler, 1981), which is much lower than the 15-25% reduction observed in our study in the air-conditioned sites.

## Neutralization

The neutralization rate for acidic aerosols can be calculated by assuming that rates of penetration and deposition of aerosol sulfate and acidity are the same, since they are associated with the same particles. Therefore, any difference in the measured indoor-outdoor ratio for the two species is due to the neutralization reaction. Starting with the simple, steady-state box model, this yields:

$$k = RP \{ [I/O(H^+)]^{-1} - [I/O(SO_4^{2-})]^{-1} \} \quad (2)$$

where  $k$  is the rate of neutralization ( $h^{-1}$ );  $P$  is the penetration efficiency (dimensionless  $0 < P < 1$ );  $R$  is the air exchange rate ( $h^{-1}$ ). The sites were modeled with a range of ventilation rates (Brauer et al., 1991; Weschler and Shields, 1989), assuming air exchange rates of 2-4 for MH and 0.5-1.0 for LN and DC. The calculated half-lives of acidic aerosols are determined in Table 7. These times for acidic aerosol

**Table 7** Neutralization rates for indoor locations

Site	Volume m <sup>3</sup>	P <sup>a</sup>	R <sup>b</sup>	Half-life min.
MH (D)	350	0.91	2-4	10-20
MH (N)	350	0.93	2-4	35-70
LN (D)	92	0.74	0.5-1.0	49-98
DC (D)	60	0.82	0.5-1.0	53-106

<sup>a</sup> penetration ratio (based on indoor/outdoor sulfate ratio)

<sup>b</sup> air exchange rate ( $h^{-1}$ )



neutralization are much longer than laboratory rates for pure sulfuric acid neutralized by ammonia – a few seconds to minutes (McMurry et al., 1983). They are, however, comparable with findings reported by Brauer et al. (1991) of approximately 15 min for Boston homes.

It is interesting to note a significantly slower neutralization rate at MH night-time than MH daytime while comparable levels of ammonia were present for both periods. Despite the large amounts of ammonia detected, acidic aerosol still penetrated indoors without being completely neutralized. Hence, ammonia levels appear to be only one of the factors affecting the indoor levels of acidic aerosols. Thus, the apparent slower rate of neutralization implies high potential risks of acidic aerosol exposures indoors during night-time periods.

### Ammonia Emissions

A box model can also be used to calculate the emission rates of indoor ammonia. Neglecting surface deposition and infiltration from outdoors (where levels are much lower), a volume source rate can be calculated:

$$E_{is} = R V C_i \quad (3)$$

where  $E_{is}$  is the emission rate ( $\mu\text{mole/h}^{-1}$ );  $C_i$  is the indoor concentration ( $\mu\text{mole/m}^{-3}$ );  $R$  is the air exchange rate ( $\text{h}^{-1}$ ); and  $V$  is the room volume ( $\text{m}^3$ ). Using the same  $R$  values in Table 7, the ammonia emission rates for the indoor sites are 800-1600, 100-200 and 75-150  $\mu\text{mole/h}^{-1}$  for MH, LN and DC, respectively.

The sources of indoor ammonia are usually associated with human emissions such as exhaled breath, sweat and the use of ammonia-based cleansers. The levels of ammonia in human breath can be very high; the range reported for oral breathing is 100 to 1000 ppb (Larson et al., 1977; Norwood et al., 1991). A median value for available data is approximately 400 ppb. The hourly ammonia exhalation rate for an individual could be 8  $\mu\text{mole h}^{-1}$ , based on an average minute ventilation rate of 8 lpm.

Human sweat is another source of ammonia indoors. This ammonia is excreted from sweat glands as urea which is hydrolyzed on dermal surfaces. The concentration of ammonia in sweat is approximately  $7 \times 10^{-5}$  by weight (Kuno, 1956). For less active individuals, the sweating rate is about 36 g/h (Consolazio et al. 1962). Hence, the hourly ammonia emission contributed from sweating could be 150  $\mu\text{mole/}$

**Table 8** Values for human ventilation rates (USEPA, 1989)

	Minute Ventilation Rate (lpm)			
	Resting	Light	Moderate	Heavy
Adult	8.3	10	35	65
Child (6-10 year)	6.7	15	43	55
Example activities	sleeping reading watching TV	housework walking	climbing stairs indoor cleaning	aerobic exercise

h at 70 °F and 40-60% relative humidity. The ammonia emission rate will be affected by heat stress. An increase in temperature not only enhances the release of sweat, but also increases the conversion rate from urea to ammonia on dermal surfaces. Based on these calculations, an individual's emission rate of ammonia from sweating may be  $\geq 20 \times$  higher than from exhaled ammonia.

An interesting question is how many people would it take to elevate indoor concentrations to the values observed in this study? For example, the LN site ( $V = 92 \text{ m}^3$ ) had mean  $\text{NH}_3$  of 56 ppb. Assuming  $R = 0.5\text{-}1.0$ , it would require only 1-2 people residing full-time in the room to serve as the aforementioned ammonia source. The calculations for MH and DC yield 6-12 and 1-2 people, respectively. These numbers are comparable, if not slightly lower than the average occupancies, supporting our contention that human-related emissions are the main indoor ammonia source.

### Dose Calculations for Study Populations

In this study, we focused our investigation on institutional settings where potentially sensitive populations (elderly persons and children) might reside or spend substantial time. The differences between indoor and outdoor acidic aerosol levels have an important impact on human exposure estimates. Population exposures will depend upon the time patterns which determine individuals' location and activities.

Among the elderly, the base rates of minute ventilation are likely to be somewhat lower than for the standard adult. Also, the periods of exertion for elderly convalescent patients would be expected to be minimal. Even mild exercise (such as a walk outdoors), however, might greatly elevate the breathing rate of a convalescing individual. The activity of children often includes a greater proportion of higher exertion levels. Inhalation dose can be characterized in the form:

$$\text{Dose} = \sum (C_i \times V_i \times T_i) \quad (4)$$

where  $C_i$  is the local concentration;  $V_i$  is the minute ventilation rate; and  $T_i$  is duration of time in micro-environment. Minute ventilation rates vary according to individuals' ages and their exertion level or activity. The average rates for an adult are given in Table 8 (USEPA, 1989b).

For acidic aerosols, the ambient concentrations represent the upper limit of exposure, since indoor sources are inconsequential. Because of the aerosol reactivity, indoor exposures for acidity are necessarily lower. Hence, human exposures depend largely upon the factors that govern indoor acidic aerosol levels (penetration and neutralization by ammonia) and the time spent outdoors.

People on average spend approximately 2-h per day outdoors and the rest of their time (92%) indoors (USEPA, 1989b). The elderly may spend upwards to 100% of their time indoors, while children may spend the maximum time, up to 10 hours per day outdoors (Spengler et al., 1989). Further, it is important to divide daily exposures into at least two distinct exposure periods, day and night. This distinction is necessary because the concentrations and activities can differ dramatically.

There are no night-time exposure measurements for the residents at LN nor night-time exposure information for children at DC when they were at home. Therefore, the night-time exposures for these populations are based on central outdoor night-time data and the calculated penetration of outdoor pollutants at the sites. For the LN site, the average night-time penetration  $[(I/O)H^+]$  was assumed to be the same as for the daytime periods (0.4). Night-time

acidities measured at RC were scaled by this penetration ratio. For the DC children exposure estimation, the night-time penetration is taken from a study in residential Boston (0.5) (Brauer et al., 1991), and night-time levels of acidity at RC were again scaled to give indoor acid exposures. We assumed that all the children who attended the day-care center lived close by, where the acidity measured at RC could adequately represent the levels near the residences.

Light and moderate ventilation rates were used for the elderly and children, respectively, during the daytime periods. Resting ventilation rates were applied for both populations during the night-time periods. The calculated doses of acidic aerosol for different sites/population are shown in Table 9. The daily doses based on average parameters were 380, 290 and 1100 nmole of acid (19, 15, 55  $\mu\text{g}$  as  $\text{H}_2\text{SO}_4$ ) for MH, LN and DC over a 24-h period, respectively.

"Worst-case" exposure values were estimated assuming higher outdoor duration, concentration of acidity, and minute ventilation rates. These assumptions include 4-h outdoor duration for elderly and children during the summer, peak concentration observed during the study period, and moderate and heavy minute ventilation rates for elderly and children, respectively.

Children at DC received doses 2 to 4 $\times$  higher than those for the elderly. This was chiefly contributed by the higher ventilation rate assumed for the children. The daily dose received by the population under study ranged from 290 to 1100 nmole of acid (15 to 55  $\mu\text{g}$  as  $\text{H}_2\text{SO}_4$ ). The "worst-case" estimate was 920 and 3400 nmole of acid (46 and 170  $\mu\text{g}$  as  $\text{H}_2\text{SO}_4$ ) for elderly and children, respectively.

**Table 9** The calculated dose of acidic aerosol at different sites/population (unit in nanomoles)

	Elderly persons		Children
	MH	LN	DC
Night-time <sup>a</sup>	135	74	75
Average daytime <sup>b</sup>	245 (64%) <sup>d</sup>	220 (75%)	1039 (93%)
"Worst case" daytime <sup>c</sup>	1074 (89%)	916 (93%)	3421 (98%)

<sup>a</sup> 12-h indoor duration and resting minute ventilation rate for both populations.

<sup>b</sup> Average 2-h outdoor duration for elderly persons at MH and LN sites; 4-h for children at DC.

<sup>c</sup> Average 4-h outdoor duration and moderate ventilation rate for the elderly at MH and LN; 4-h outdoor duration and heavy ventilation rate were applied for children at DC. Peak concentrations of acidity were used for microenvironments indoor/outdoor.

<sup>d</sup> Percentage of total daily dose from daytime period.

### Comparison of Exposure Dose

It is meaningful to compare these estimates to the doses experienced by subjects in several recent studies of acidic aerosol exposure. In two recent clinical studies, pulmonary function changes were associated with short-term exposures to acidic sulfate aerosols at concentration 100 and 450  $\mu\text{g}/\text{m}^3$  with duration 40 and 10 minutes, respectively (Koenig et al., 1989 and Utell et al., 1983). The delivered doses calculated by Spengler et al. (1989) for the aforementioned studies are 1300 and 1200 nmoles of acidic sulfate aerosols. In the field, effects for winter-time exposures to acidic aerosols were reported for a study of 207 asthmatics in Denver by Ostro et al. (1991). From daily data for respiratory symptoms,

cough and shortness of breath were found to be significantly correlated with exposure-adjusted  $H^+$  concentrations, with a dose-response relationship observed in the range from 25 to 500 nmoles. This range was calculated based on the combination of outdoor concentrations and exposure pattern (8-h indoor resting, 1-h outdoor exercise, 1-h moderate outdoor activities and 14-h indoor light activities). The indoor-outdoor penetration rate was assumed 0.5 for all calculations of indoor environments.

In the present study, doses received by the elderly and children ranged from 290 to 1100 nmoles of acid (15-55  $\mu\text{g}$  as  $H_2SO_4$ ) in a 24-h period, and the "worst-case" dose received by children in the daytime could be as high as 3400 nmoles of acid. Our findings were consistent with the dose calculations by Spengler et al. (1989) where observed doses were comparable to the clinical and epidemiological studies involving resultant health effects. The potential existed for the populations in our study to experience some respiratory effects.

## Conclusions

At the three institutional settings investigated, high concentrations of acidic sulfate aerosols, nitric acid and ozone were measured during the daytime monitoring periods. For most of the pollutant species measured, the major source of indoor contamination was infiltration from the outdoor environment, except ammonia and nitrate aerosols which seemed derive from indoor sources.

Indoor concentrations of pollutants were driven by outdoor levels. Based on the indoor-to-outdoor (I/O) sulfate ratios, the penetration of fine aerosol was >70% at the three institutional settings. The penetration for closed-circulation ventilation locations was 15-25% lower than for an open-window setting. The I/O ratio for aerosol acidity was 0.4 to 0.5, indicating that substantial neutralization took place indoors. Calculation of neutralization rates indicated that the mean reaction half lives for acidic aerosols was between 10 and 106 minutes.

Indoor levels of ammonia were  $\geq 10\times$  higher than corresponding outdoor values. Indoor ammonia levels were consistent with the emission rates calculated to be due mainly to human occupants. However, the high ammonia levels only partially neutralized acidic aerosols penetrating from outdoors. Aerosol ammonium nitrate formation was found to be more favorable in air-conditioned buildings than in open ventilation settings due to the

lower indoor temperature. Due to substantial loss of nitric acid during penetration as well as to low indoor nitrate levels (smaller than detection limit), the ammonium nitrate system could not achieve equilibrium in half of the sampling periods. The agreement between theoretical and measured values of the ammonia-nitric acid partial pressure product was poor.

The magnitude of indoor exposure and dose depends upon outdoor penetration and neutralization by indoor ammonia. Depending upon activity patterns and ventilation rates, doses received by different populations may differ substantially. Children may receive more than 93% of their dose from outdoor exposure due to their outdoor activities in the summer. The daily dose received by the population under study ranged from 290 to 1100 nmole of acid (15 to 55  $\mu\text{g}$  as  $H_2SO_4$ ). The worst case estimate was 920 and 3400 nmole of acid (46 and 170  $\mu\text{g}$  as  $H_2SO_4$ ) for elderly persons and children, respectively. The dose of acid delivered to children was calculated to be 2 to 4 $\times$  higher than the dose to elderly populations over a 24-h period. This suggests that, due to their activity pattern, children may be at greater risk from summertime acidic aerosol exposure. Furthermore, the calculated doses on episode days was comparable to the doses which led to respiratory responses in clinical studies.

## Acknowledgements

This study was sponsored by the New Jersey Department of Environmental Protection Agency/Division of Science and Research. Additional funding was provided by the US Environmental Protection Agency/Health Effects Research Laboratory as part of the NIEHS Center of Excellence (ES05022). Fellowship support for the author, Chris S.K. Liang, was generously provided by EOHSI and Cook College. We would like to thank Dr. Premlata Menon for her support in the field, suggestions in the laboratory and kindness in many ways. We would like to acknowledge the assistance of David Morris and Lung-Cheng Hwang in their field and laboratory work.

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