

## MEASUREMENT AND MODELING OF AIRBORNE CONCENTRATIONS AND INDOOR SURFACE ACCUMULATION RATES OF IONIC SUBSTANCES AT NEENAH, WISCONSIN

J. D. SINCLAIR and L. A. PSOTA-KELTY

AT&T Bell Laboratories, 600 Mountain Avenue, Murray Hill, NJ 07974-2070, U.S.A.

and

C. J. WESCHLER and H. C. SHIELDS

Bell Communications Research, 331 Newman Springs Road, Red Bank, NJ 07701-7020, U.S.A.

(First received 10 January 1989 and in final form 22 September 1989)

**Abstract**—The objective of this continuing investigation of indoor/outdoor/surface relationships has been to develop an accurate method for predicting and subsequently managing the accumulation rates and ultimately the effects of corrosive substances on electronic equipment surfaces in field and manufacturing environments. We previously reported indoor/outdoor ratios and deposition velocities for  $\text{Cl}^-$ ,  $\text{SO}_4^{2-}$ ,  $\text{Na}^+$ ,  $\text{NH}_4^+$ ,  $\text{K}^+$ ,  $\text{Mg}^{2+}$  and  $\text{Ca}^{2+}$  associated with fine and coarse particles at telephone company switching equipment locations in Wichita (Kansas), Lubbock (Texas) and Newark (New Jersey). Using the results from these studies, a methodology was developed for predicting the average indoor surface accumulation rates of ionic substances from their outdoor concentrations.

In this paper we report new results for a site at Neenah, Wisconsin. At this site detailed data on the operational status of the air handling equipment were also obtained through a permanent monitoring system. These data and the data on ionic species have been used in a mass balance model that calculates indoor concentrations from outdoor concentrations. Coupling this mass balance model with the measured deposition velocities substantially improves the earlier methodology for predicting surface accumulation rates from outdoor concentrations and enables decision makers to evaluate the effects of various manipulations in critical air handling system operating variables. Informed decisions can now be made when striking a balance between energy use and indoor air quality or equipment reliability.

**Key word index:** Indoor air quality, deposition velocities, particulate contamination, soiling, acidic aerosols, mass balance model, sulfate, ammonium, chloride, fine particles, coarse particles, indoor/outdoor ratios.

### INTRODUCTION

In earlier work (Sinclair *et al.*, 1985, 1988a) extensive data on outdoor and indoor airborne concentrations of water soluble ionic substances at electronic equipment locations in Wichita (Kansas), Lubbock (Texas) and Newark (New Jersey) were presented. The accumulation rates of these substances on the equipment surfaces were also determined from water extracts (Sinclair and Psota-Kelty, 1984) collected from surfaces that were undisturbed for periods ranging from 7 to 15 years. Airborne samples were obtained using dichotomous samplers located within the equipment rooms and on the roofs of these buildings. Samples were collected on Teflon filters and then analyzed, after water extraction, by ion chromatography. Surface samples were collected by filter paper extraction (Sinclair, 1982) and then analyzed in a manner similar to that used for the Teflon filters. From these data, indoor/outdoor (*I/O*) ratios and deposition velocities were determined for the ions associated with fine and coarse particles.

The *I/O* ratios when building fans are activated, for ions that do not have indoor sources, can be approximated by a relationship given by Weschler *et al.* (1983) that requires as inputs: (1) the fraction of air leaking into and out of the building; (2) the fraction of recirculation made up with outside air; (3) the fractional filter efficiency of leakage paths; (4) the dust deposition velocity; (5) the internal dust deposition area; (6) the volume of air flow in the air handling system; and (7) the fraction of particles in a given size range removed by the building filters. Prior to this continuing study of indoor/outdoor/surface relationships, all of the inputs were well known or could be approximated except the deposition velocity. Our preliminary data for coarse particles indicated that, at the air flow velocities present at these locations (typically in the range of  $1\text{--}6\text{ m min}^{-1}$ ), the average deposition velocity is roughly what would be predicted for a deposition process dominated by gravitational settling. For fine particles, the average deposition velocity is larger than that predicted by gravitational settling alone. This discrepancy reflects the import-

Table 1. Airborne concentrations for ionic species and total mass associated with fine and coarse particles ( $\text{ng m}^{-3}$ )

Sample	Size	Mass	$\text{SO}_4^{2-}$	$\text{Cl}^-$	$\text{Na}^+$	$\text{NH}_4^+$	$\text{K}^+$	$\text{Mg}^{2+}$	$\text{Ca}^{2+}$
2-7/4									
Indoor	Fine	1220	675	2	11	152	0	2	9
	Coarse	1500	75	2	3	5	4	4	17
Outdoor	Fine	13,730	3101	49	47	1647	25	25	45
	Coarse	13,790	564	69	36	0	20	271	726
15-22/4									
Indoor	Fine	1220	616	0	5	173	5	2	5
	Coarse	NA*	17	3	2	0	2	3	4
Outdoor	Fine	10,330	3715	3	51	1188	53	18	48
	Coarse	13,380	307	31	37	0	14	245	626
29/4-5/5									
Indoor	Fine	670	460	4	6	152	8	2	4
	Coarse	660	24	1	3	0	0	1	5
Outdoor	Fine	7800	2270	7	36	749	47	25	56
	Coarse	24,580	148	41	43	1	31	361	910
19-26/5									
Indoor	Fine	2410	1090	0	7	363	7	2	5
	Coarse	780	25	0	2	1	0	1	2
Outdoor	Fine	12,400	5158	3	44	1491	56	21	64
	Coarse	18,630	640	34	34	0	21	221	669
9-16/6									
Indoor	Fine	2580	953	0	8	292	7	2	7
	Coarse	380	37	0	2	5	0	1	4
Outdoor	Fine	13,940	4706	3	45	1597	71	30	117
	Coarse	16,350	540	47	24	0	39	387	1732
7-14/7									
Indoor	Fine	1530	451	0	5	154	5	2	3
	Coarse	160	23	0	1	1	0	1	1
Outdoor	Fine	8640	2592	3	28	841	45	18	45
	Coarse	8170	221	10	15	0	18	235	616
5-12/8									
Indoor	Fine	2050	745	3	10	222	13	3	6
	Coarse	180	13	0	1	2	0	0	0
Outdoor	Fine	11,080	3976	2	46	1260	82	25	83
	Coarse	4890	121	4	9	0	0	137	361
19-25/8									
Indoor	Fine	1800	581	2	6	180	4	2	10
	Coarse	360	60	1	3	3	1	0	1
Outdoor	Fine	11,550	—	—	—	—	—	—	—
	Coarse	3240	—	—	—	—	—	—	—
1-8/9									
Indoor	Fine	2250	1298	1	8	400	11	1	2
	Coarse	90	6	1	2	0	2	2	12
Outdoor	Fine	13,910	7124	3	28	2065	91	23	59
	Coarse	4200	14	6	12	0	12	160	408
23-30/9									
Indoor	Fine	1540	526	2	5	157	5	2	3
	Coarse	60	40	1	1	6	0	0	1
Outdoor	Fine	11,570	3327	23	31	1024	66	45	109
	Coarse	1420	97	4	7	0	2	48	95
7-14/10									
Indoor	Fine	1530	662	2	6	251	3	0	0
	Coarse	160	13	0	1	0	0	1	1
Outdoor	Fine	9220	2568	23	20	1100	45	38	127
	Coarse	1030	23	5	2	0	0	32	81

Table 1. (Contd.)

Sample	Size	Mass	SO <sub>4</sub> <sup>2-</sup>	Cl <sup>-</sup>	Na <sup>+</sup>	NH <sub>4</sub> <sup>+</sup>	K <sup>+</sup>	Mg <sup>2+</sup>	Ca <sup>2+</sup>
21-27/10									
Indoor	Fine	3230	1196	7	11	361	20	4	3
	Coarse	50	22	2	1	2	0	1	2
Outdoor	Fine	19,420	3462	422	44	2124	127	50	121
	Coarse	1770	222	4	6	0	0	56	133
27/10-3/11									
Indoor	Fine	1760	424	2	12	156	21	3	2
	Coarse	330	70	1	1	7	3	1	3
Outdoor	Fine	12,120	2170	40	38	1014	130	56	94
	Coarse	1400	34	8	7	0	0	52	107
17-24/11									
Indoor	Fine	1300	375	2	3	119	8	2	3
	Coarse	NA	43	1	4	9	4	1	2
Outdoor	Fine	14,450	3185	188	40	1150	107	21	31
	Coarse	1950	113	26	23	132	3	48	111
15-22/12									
Indoor	Fine	1020	305	3	5	98	17	3	2
	Coarse	NA	43	2	1	1	2	1	3
Outdoor	Fine	21,630	3406	233	51	2636	142	28	17
	Coarse	1130	217	44	41	64	0	18	43
5-12/1									
Indoor	Fine	960	287	2	5	96	17	2	2
	Coarse	40	13	3	8	2	1	1	4
Outdoor	Fine	33,270	6603	623	59	5980	35	12	23
	Coarse	1120	514	0	47	122	18	9	19
26/1-2/2									
Indoor	Fine	700	254	1	7	67	6	2	5
	Coarse	200	22	3	13	5	0	2	5
Outdoor	Fine	15,470	2935	112	165	2152	50	23	43
	Coarse	1030	65	126	76	0	0	42	112
16-23/2									
Indoor	Fine	1110	296	1	4	125	8	8	21
	Coarse	480	37	20	13	2	4	12	34
Outdoor	Fine	23,140	2935	307	74	3251	135	44	71
	Coarse	NA	65	0	10	0	0	16	43

\*Not available due to large uncertainty associated with dichotomous sampler mass measurements when the fine particle concentration is very high and the coarse particle concentration is very low

the outdoor coarse concentration, while for calcium the outdoor coarse concentration is commonly much larger than the outdoor fine. The outdoor Ca<sup>2+</sup> concentration in coarse particles shows a wide range (19–1732 ng m<sup>-3</sup>) while the range of the outdoor calcium in fine particles is narrow (17–127 ng m<sup>-3</sup>), so that for a few intervals the outdoor fine is larger than the outdoor coarse concentration. The highest masses and highest Ca<sup>2+</sup> concentrations for outdoor coarse particles occur in the spring and early summer, probably reflecting farming activities. The lowest concentrations occur in the winter, when snow cover reduces the contribution of soil-derived particles. The masses and ion concentrations for outdoor fine particles are highest during the heating season (fossil fuel combustion) and lowest during the spring and early fall.

For SO<sub>4</sub><sup>2-</sup> concentrations in fine mode particles, the indoor concentrations tend to track the outdoor concentrations. This is illustrated in Fig. 1 for representative periods throughout this study. The tracking is not as good during the winter months when very little outside air is brought into the building. The NH<sub>4</sub><sup>+</sup> concentrations (from Table 1) show a similar tracking pattern to that observed for SO<sub>4</sub><sup>2-</sup>. The behavior for Ca<sup>2+</sup> was dramatically different. The indoor concentration did not track the outdoor concentration. These observations can be explained by consideration of the efficiencies of the filters used in the building's air handling system (see Experimental section). The mass fraction of coarse particles removed by these filters is approximately 0.995, while the mass fraction of fine particles removed is approximately 0.75–0.85 (Bauer

Table 6 contrasts the weekly status of the air handling system with the weekly *I/O* ratios for fine particles and fine mode sulfate. The *I/O* ratios for both particles and  $\text{SO}_4^{2-}$  are lowest from 15 December to 23 February, ranging from 0.03 to 0.10. During this period the amount of outside air brought into the building was at a minimum. Further comparisons between the HVAC parameters and the corresponding *I/O* ratios in Table 6 suggest that the *I/O* ratio varies with both the amount of outside air introduced and the fraction of time the building fans are operated. This conclusion is supported by the larger data set on fine particle concentrations from 25 March 1986 to 27 July 1987 (Weschler and Shields, 1989). Introduction of larger amounts of outside air produces larger *I/O* ratios. Increased use of building fans produces smaller *I/O* ratios because of recirculation through the air filters.

#### Mass-balance model

The influence of the various air handling system operating parameters on the steady-state concentration of the indoor airborne particles can be examined in the context of a mass-balance model (Weschler *et al.*, 1988; Weschler and Shields, 1989). The mass balance equation is

$$C_i = (x) \left[ \frac{R_i + v_1(1-F_1)C_e + v^*(1-F_p)(1-F_s)fC_e}{k_d A_d + v^* F_s(1-f) + v_1 + v^* f} \right] + (1-x) \left[ \frac{R_i + v'_1(1-F_1)C_e}{k_d A_d + v'_1} \right] \quad (1)$$

where:  $C_i$  = steady-state indoor concentration of the species ( $\mu\text{g m}^{-3}$ );  $C_e$  = average outdoor concentration of the species ( $\mu\text{g m}^{-3}$ );  $x$  = fraction of time the building fans are on (0–1);  $f$  = fraction of recirculation made up with outside air (0–1);  $R_i$  = rate at which airborne

ionic substances are generated indoors ( $\mu\text{g min}^{-1}$ );  $v_1$  = volumetric flow rate for air leaking into and out of the building when the building fans are operating ( $\text{m}^3 \text{min}^{-1}$ );  $v'_1$  = volumetric flow rate for air leaking into and out of the building when the building fans are not operating ( $\text{m}^3 \text{min}^{-1}$ );  $v^*$  = volumetric flow rate for air in the air handling system ( $\text{m}^3 \text{min}^{-1}$ ) (the range of the variable volume system at Neenah was 34–184  $\text{m}^3 \text{min}^{-1}$  and, to a first approximation, a typical value was 170  $\text{m}^3 \text{min}^{-1}$ , based on values observed throughout the year);  $F_1$  = fractional equivalent filter efficiency of the leakage paths (0–1), which is approximately 0.2 for ions associated with fine particles and 1 for ions in coarse particles;  $F_p$  = fractional primary filter efficiency (equal to 0 at Neenah, since there are no primary filters);  $F_s$  = fractional secondary filter efficiency (for ions associated with fine particles a value between 0.75 and 0.85 is assumed and for ions in coarse particles a value of 0.995 is assumed (Bauer *et al.*, 1973));  $k_d$  = internal deposition velocity of the species ( $\text{m min}^{-1}$ ), based on measurements at Neenah and other sites (Sinclair *et al.*, 1988a and this work), with the values used being 0.0024–0.036  $\text{m min}^{-1}$  for ions associated with fine particles and 0.4  $\text{m min}^{-1}$  for ions associated with coarse particles (see below);  $A_d$  = internal deposition area of the species ( $\text{m}^2$ ), which is assumed equal to the floor area of 715  $\text{m}^2$  for the coarse particles and 5000  $\text{m}^2$  for fine particles (this value is normally 5–15 times the floor area, since fine particles deposit on top-side horizontal, bottom-side horizontal, and vertical surfaces).

Using this model along with the specified values for the model parameters, the values from Table 2 for the 'Fraction of time fans on' and 'Fraction of outside air', and the values from Table 1 for the outdoor concentrations, steady-state indoor concentrations can be calculated that are in reasonable agreement with the actual measured values reported in Table 1. The most

Table 6. Status of air handling system and corresponding indoor/outdoor ratios for fine particles and fine mode  $\text{SO}_4^{2-}$

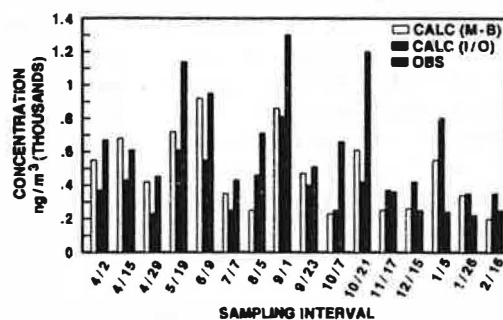
Week	Fraction of time fan on	Fraction of outside air	<i>I/O</i> Fine particles	<i>I/O</i> $\text{SO}_4^{2-}$
1–7/4	0.87	0.64	0.09	0.22
15–22/4	0.89	0.67	0.12	0.17
29/4–5/5	0.96	0.74	0.09	0.20
19–26/5	0.90	0.50	0.19	0.21
9–16/6	0.88	0.75	0.18	0.20
7–14/7	0.91	0.46	0.18	0.17
5–12/8	0.99	0.26	0.19	0.19
19–25/8	0.90	0.62	0.16	NA
1–8/9	0.92	0.40	0.16	0.18
23–30/9	0.87	0.46	0.13	0.16
7–14/10	0.96	0.34	0.16	0.26
21–27/10	0.98	0.70	0.17	0.35
17–24/11	1.0	0.36	0.09	0.12
15–22/12	1.0	0.34	0.05	0.09
5–12/1	1.0	0.31	0.03	0.04
26/1–2/2	0.93	0.38	0.05	0.09
16–23/2	1.0	0.27	0.05	0.10

Table 7. Calculated vs observed concentrations of  $\text{SO}_4^{2-}$  and  $\text{Ca}^{2+}$  ions associated with fine particles at Neenah, WI ( $\mu\text{g m}^{-3}$ )

Week	$\text{SO}_4^{2-}$ * (calculated)	$\text{SO}_4^{2-}$ (observed)	$\text{Ca}^{2+}$ † (calculated)	$\text{Ca}^{2+}$ (observed)
2-7/4	559	675	2	9
15-22/4	681	616	2	5
29/4-5/5	428	460	3	4
19-26/5	765	1090	2	5
9-16/6	936	953	6	7
7-14/7	359	451	2	3
5-12/8	301	745	2	6
1-8/9	880	1298	2	2
23-30/9	483	526	4	3
7-14/10	262	662	3	0
21-27/10	617	1196	6	3
17-24/11	315	375	1	3
15-22/12	320	305	0.5	2
5-12/1	570	287	0.5	2
26/1-2/2	344	254	1	5
16-23/2	223	296	2	21

\*Calculated using  $k_d = 0.004 \text{ cm s}^{-1} = 0.0024 \text{ m min}^{-1}$ ;  $F_s = 0.75$ .†Calculated using  $k_d = 0.06 \text{ cm s}^{-1} = 0.0036 \text{ m min}^{-1}$ ;  $F_s = 0.85$ .

significant unknown in the model is  $R_i$ , the rate at which particles are generated within the office. For fine particles, where chemical analyses suggest that a large fraction of these particles have infiltrated from outdoors,  $R_i$  can be initially approximated by a small non-zero value (e.g.  $50\text{--}100 \mu\text{g min}^{-1}$ —this corresponds to about 12 cigarettes  $\text{day}^{-1}$ ). For a few ions associated with fine particles, an even smaller value of  $R_i$  is appropriate. Table 7 compares calculated steady-state indoor concentrations for fine-mode  $\text{SO}_4^{2-}$  and fine-mode  $\text{Ca}^{2+}$  with the observed concentrations for these ions. The value of  $R_i$  used in these calculations was zero. Different values for  $k_d$  and  $F_s$  were used in the  $\text{SO}_4^{2-}$  and  $\text{Ca}^{2+}$  calculations, reflecting the different mass median diameters within the fine mode. A zero value is a reasonable approximation for  $\text{SO}_4^{2-}$ ; no significant indoor sources of fine-mode  $\text{SO}_4^{2-}$  have been identified at this site. For  $\text{Ca}^{2+}$ , this situation is more complicated. Humidification during dry winter months can generate small amounts of fine-mode  $\text{Ca}^{2+}$ . Examining the data in Table 7 reveals reasonable agreement between fine-mode  $\text{SO}_4^{2-}$  values calculated with the mass-balance model and those actually observed. For  $\text{Ca}^{2+}$ , the agreement is not poor, but can be significantly improved by using a non-zero value for  $R_i$  of  $1 \mu\text{g min}^{-1}$ . This is a very small value (compared with  $50\text{--}100 \mu\text{g min}^{-1}$  for 12 cigarettes  $\text{day}^{-1}$ ) and is a reasonable approximation for the Neenah site. For other ions with known indoor sources (e.g. K), the mass-balance model can be used to estimate the internal emission rate responsible for the observed indoor concentrations. Figure 2 compares indoor fine-mode  $\text{SO}_4^{2-}$  values calculated using the mass-balance model and those calculated using the more empirical approach, based on  $I/O$  ratios, with the actual values measured at the Neenah site. For all but three sampling periods, the mass-balance model

Fig. 2. Comparison of calculated and observed indoor concentrations of fine mode  $\text{SO}_4^{2-}$  for 7 day sampling intervals beginning on the indicated dates.

produces a better (or comparable) prediction of the indoor concentrations. The conditions represented in Table 7 and Fig. 2 span all four seasons and their weather extremes, as well as periods when maximum outside air, minimum outside air, maximum fan operation, or minimum fan operation were in effect.

#### Surface ionic species

The average accumulation rates of ions on surfaces at the Neenah location, as shown in Table 3, were considerably greater on top-side horizontal surfaces than on vertical surfaces. This result reflects the fact that top-side horizontal surfaces accumulate both fine and coarse particles, whereas vertical surfaces accumulate primarily fine particles (Sinclair *et al.*, 1985). The largest contributor to accumulation on vertical surfaces is  $\text{SO}_4^{2-}$ , consistent with its being the major component of fine particles. The similar accumulation rates of  $\text{Cl}^-$  on Zn and Al surfaces is consistent with other midwestern locations and contrasts with north-eastern locations where preferential accumulation on

Table 8. Source contributions to average ion accumulation rates on surfaces at Neenah ( $\mu\text{g cm}^{-2}$ )

	Gas induced reaction	Coarse particle deposition	Fine particle deposition
$\text{SO}_4^{2-}$		0.15	0.07
$\text{Cl}^-$	-0.01	0.06	0.03
$\text{NO}_3^-$		0.28	0.02
$\text{Na}^+$		0.04	0.02
$\text{K}^+$		0.04	0
$\text{Mg}^{2+}$		0.05	0.005
$\text{Ca}^{2+}$		0.41	0.01

Zn surfaces was observed. In the midwest the accumulation is dominated by particle deposition, whereas in the northeast corrosive Cl containing gases cause preferential accumulation on Zn surfaces.

The average accumulation rates given in Table 3 for horizontal and vertical Zn and Al surfaces can be converted to accumulations due to each of the three possible major sources of each ion, namely fine particles, coarse particles, and, for  $\text{Cl}^-$  at appropriate locations, reactive gases (Sinclair *et al.*, 1985). The calculated contributions from each of the sources for each ion except  $\text{NH}_4^+$  are given in Table 8. The behavior of  $\text{NH}_4^+$  is unusual in that the  $\text{NH}_4^+$  concentrations on Zn and Al reach a nearly constant value after very short exposure to the ambient. Possible mechanisms for  $\text{NH}_3$  loss are discussed elsewhere (Sinclair *et al.*, 1985).

#### Deposition velocities

Average deposition velocities for ions in coarse and fine particles at Neenah are presented in Table 9 along with the previously obtained data for Wichita, Lubbock and Newark. As expected, the gravitational settling velocity for a typical coarse particle ( $0.6 \text{ cm s}^{-1}$  for a  $10 \mu\text{m}$  particle of  $\text{CaCO}_3$ ) is within the range of deposition velocities measured for ions in the coarse mode at all four locations. The first results, which came from Wichita and Lubbock, exhibited deposition velocities for the various ions in the coarse particles that spanned a substantial range. This range was thought to be due to the short sampling period of these studies. At Newark a substantial amount of data were obtained and the observed variation was very small. Surprisingly, at Neenah, where a substantial amount of data has also been obtained, the deposition velocities span a larger range than seen at Newark. This range may be partly a result of the large uncertainty in the concentrations of ions associated with coarse particles. However, averages based on 11 months of sampling are likely to smooth out these variations. While both the Newark and Neenah locations are served by high efficiency filters, the average size of the coarse particles at Neenah appears to be larger and the composition appears to vary with size. The larger particles in the coarse particle fraction appear to be calcium rich while the smaller particles in the coarse mode appear

Table 9. Comparison of deposition velocities for ions associated with fine and coarse particles ( $\text{cm s}^{-1}$ )

	Neenah	Newark	Wichita	Lubbock
Fine particles				
$\text{Cl}^-$	0.5	<0.01	0.03	<0.01
$\text{SO}_4^{2-}$	0.004	0.005	0.004	0.005
$\text{Na}^+$	0.09	0.05	0.2	0.07
$\text{K}^+$	0	0.008	0.004	0.03
$\text{Mg}^{2+}$	0.08	0.02	<0.01	0.03
$\text{Ca}^{2+}$	0.06	0.03	<0.02	0.006
Coarse particles				
$\text{Cl}^-$	0.9	0.2	0.8	0.2
$\text{SO}_4^{2-}$	0.1	0.09	1.8	0.1
$\text{Na}^+$	0.4	0.2	0.7	0.2
$\text{K}^+$	1.3	0.3	3.8	0.2
$\text{Mg}^{2+}$	0.8	0.3	0.9	0.2
$\text{Ca}^{2+}$	2.2	0.2	1.0	0.07

to be sulfate rich. This explanation is consistent with the large  $\text{SO}_4^{2-}$  concentration in the fine mode fraction and the expected tail in the size distribution of these combustion derived particles into the coarse particle fraction. An indoor source of large  $\text{Ca}^{2+}$  rich particles may also be contributing to the large range in deposition velocities. For fine particles the variations in deposition velocities are substantial at all four locations. In view of the large amount of data collected at both Newark and Neenah, the variation must be real. Again, variation in composition over the range of sizes within the fine particle fraction can explain the observations. Calcium and  $\text{Mg}^{2+}$  are predominantly present in coarse particles. A tail in their size distribution that extends into the fine particle range would bias the size distribution of these ions in fine particles to larger sizes than predominantly fine mode ions and thus would result in higher deposition velocities than those found for  $\text{SO}_4^{2-}$ . This explanation is supported in two extensive review articles by Milford and Davidson (1985, 1987). From their work,  $\text{SO}_4^{2-}$  particles from continental sites have mass median diameters (MMDs) averaging  $0.52 \mu\text{m}$ . In contrast, the average MMDs for  $\text{Mg}^{2+}$ ,  $\text{Ca}^{2+}$  and  $\text{Na}^+$  are 6.34, 4.64 and  $3.78 \mu\text{m}$ , respectively. Further support for this explanation for the variation in deposition velocities comes

from the average  $I/O$  ratios for fine-mode species (Table 5). Figure 3 compares the  $I/O$  ratios for fine-mode particles with those for fine-mode  $\text{SO}_4^{2-}$ . The latter are, with one exception, always larger than the former. This observation indicates that fine-mode  $\text{SO}_4^{2-}$  has a smaller MMD than the average MMD for fine-mode particles. Thus, at least one component other than  $\text{SO}_4^{2-}$  must have a larger MMD than the average MMD for fine-mode particles. Fine-mode  $\text{Ca}^{2+}$  appears to be such a species. Figure 4 compares the  $I/O$  ratios for fine-mode particles with those for fine-mode  $\text{Ca}^{2+}$ . In most cases the latter are smaller than the former, indicating a larger MMD for fine-mode  $\text{Ca}^{2+}$  (exceptions are likely for periods with indoor sources of fine  $\text{Ca}^{2+}$ ). The contrast between  $\text{SO}_4^{2-}$  values in Fig. 3 and  $\text{Ca}^{2+}$  values in Fig. 4 is consistent with the contrast in deposition velocities between these two fine-mode components ( $0.004 \text{ cm s}^{-1}$  vs  $0.06 \text{ cm s}^{-1}$ ).

All of the deposition velocities obtained at Neenah and Newark for ions associated with fine particles, and most of those obtained at Wichita and Lubbock, are one or two orders of magnitude smaller than those typically found outdoors (Everett *et al.*, 1979; Wesely *et al.*, 1977; Sievering, 1982). This observation is consistent with the low air flow and reduced turbulence of an indoor environment. Outdoors, it might be expected that fine particles would usually follow turbulent eddies and that these forces would dominate

the deposition of fine particles. However, indoors such weather driven forces should be considerably less important. Thermophoretic drift, convective diffusion, electrostatic forces, and gravitational settling become more important contributors to the deposition process, and appear in many situations, based on the modeling of Nazaroff and Cass (1987), to be of comparable magnitude for fine particles. The calculated gravitational settling rate for a  $0.5 \mu\text{m}$   $\text{NH}_4\text{HSO}_4$  particle ( $0.002 \text{ cm s}^{-1}$ ), is at the low end of the range of observed deposition velocities. Variations in deposition velocities at different equipment locations within the Neenah office were attributed to variations in air currents (turbulence) near the surfaces of the equipment (Sinclair *et al.*, 1988b).

#### Predictive methodology

From the relationships observed in this study on outdoor, indoor, and surface concentrations of  $\text{NH}_4^+$ ,  $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$ ,  $\text{Ca}^{2+}$ , and other species, an estimate of the loading on indoor surfaces in the absence of indoor sources can be made for all areas where outdoor concentrations are known. Using a methodology described in detail elsewhere (Sinclair and Weschler, 1985), typical surface accumulation rates of  $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$  and  $\text{NH}_4^+$  at well sealed office buildings can be estimated from appropriate indoor/outdoor ratios and deposition velocities. Data on outdoor concentrations and building filter efficiencies are the only input needed to predict indoor surface accumulation rates. In the earlier reports on this methodology, representative values of indoor and outdoor concentrations, the ratios between these, and measured accumulation rates (most of the  $I/O$  ratio and accumulation rate data being those of the authors) were assembled to illustrate the methodology. With the availability of more extensive data from the current study at Neenah, improvements in some of the  $I/O$  ratios and deposition velocities can be made. Even more importantly, greater confidence can now be placed in the use of all of these values. Appropriate  $I/O$  ratios to use for predictions of indoor concentrations from outdoor concentrations in the absence of indoor sources are as follows: 0.3 and 0.12 for fine particles and 0.05 and 0.03 for coarse particles in buildings using standard and high efficiency filters, respectively. Table 10 provides best estimates of deposition velocities appropriate for office locations with linear flow rates averaging roughly  $2 \text{ m min}^{-1}$  with a range of  $1\text{--}6 \text{ m min}^{-1}$ . This methodology provides estimates of the average accumulation rates on horizontal or vertical surfaces across an equipment room. The range in deposition velocities and accumulation rates at each location is likely to be at least  $\pm 50$  per cent of the average, as demonstrated in related studies (Sinclair *et al.*, 1988b).

The accuracy of predictions of surface accumulation rates can be substantially improved by using indoor concentrations calculated with the mass-balance model (Equation 1). More significant than the improved

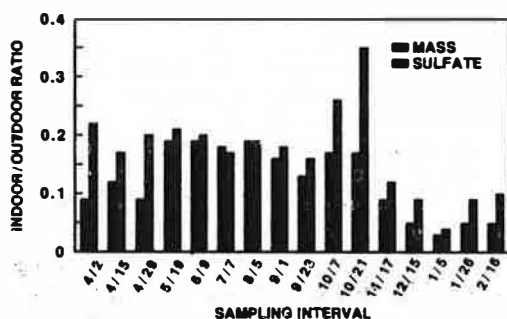


Fig. 3. Indoor/outdoor ratios for fine particles and fine  $\text{SO}_4^{2-}$  for 7 day sampling intervals beginning on the indicated dates.

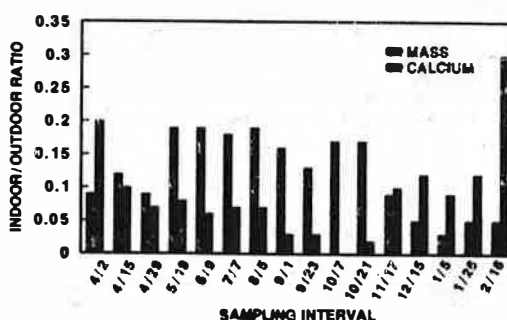


Fig. 4. Indoor/outdoor ratios for fine particles and fine  $\text{Ca}^{2+}$  for 7 day sampling intervals beginning on the indicated dates.

Table 10. Estimated indoor deposition velocities ( $\text{cm s}^{-1}$ )

Species	Velocity
Fine particles	
$\text{Cl}^-$	0.005*
$\text{SO}_4^{2-}$	0.005
$\text{Na}^+$	0.07
$\text{K}^+$	0.005*
$\text{Mg}^{2+}$	0.04
$\text{Ca}^{2+}$	0.04
Coarse particles	
All ions	0.7*

\* Very rough estimate; the available data span a range of more than an order of magnitude.

accuracy is the predictive capability inherent in this method. Critical HVAC operating parameters can be varied in the model, and the resulting effects on average accumulation rates can be evaluated. The HVAC parameters most easily changed in a commercial office building are: fraction of time the building fans are on,  $x$ ; fan speed,  $v^*$ ; primary filter efficiency,  $F_p$ ; secondary filter efficiency,  $F_s$ ; and fraction of outside air,  $f$ . Equation 1 can be used to calculate steady-state indoor concentrations that would result from different values for these parameters. The accumulation rates are directly proportional to indoor concentrations. With such a procedure, indoor particulate accumulation rates can be reduced using an informed, cost effective strategy.

### CONCLUSIONS

The objective of this continuing investigation of indoor/outdoor/surface relationships has been to develop an accurate method for predicting and subsequently managing the accumulation rates and ultimately the effects of corrosive substances and other contaminants on electronic equipment surfaces. Through the investigation at Neenah, a better understanding of the effects of the HVAC operating parameters on surface accumulation rates of water soluble pollutants has been achieved. The indoor surface accumulation rates of ions in fine particles, which includes most of the corrosive substances of concern, vary in a predictable way with the air handling system parameters. Coupling quantitative information on these parameters with deposition velocities derived from data obtained at four locations across the U.S. is sufficient to make accurate predictions of surface accumulation rates. In the absence of quantitative information on HVAC parameters, somewhat less accurate predictions of surface accumulation rates can be made based on typical I/O ratios of water soluble inorganic ions that have been measured at Wichita, Lubbock, Newark and Neenah.

### REFERENCES

- Bauer E. J., Reager B. T. and Russell C. A. (1973) Use of particle counts for filter evaluation. *ASHRAE J.* 13, 53–58.
- Everett R. G., Hicks B. B., Berg W. W. and Winchester J. W. (1979) An analysis of particulate sulfur and lead gradient data collected at Argonne National Laboratory. *Atmospheric Environment* 13, 931–934.
- Milford J. B. and Davidson C. I. (1985) The sizes of particulate trace elements in the atmosphere—a review. *J. Air Pollut. Control. Ass.* 35, 1249–1260.
- Milford J. B. and Davidson C. L. (1987) The size of sulfate and nitrate aerosols—a review. *J. Air Pollut. Control Ass.* 37, 125–134.
- Nazaroff W. W. and Cass G. R. (1987) Particle deposition from a natural convection flow onto a vertical isothermal flat plate. *J. Aerosol Sci.* 18, 445–455.
- Sievering H. (1982) Profile measurements of particle transfer at an air/land interface. *Atmospheric Environment* 16, 301–306.
- Sinclair J. D. (1982) Paper extraction for sampling inorganic salts on surfaces. *Anal. Chem.* 54, 1529–1533.
- Sinclair J. D. and Psota-Kelty L. A. (1984) Ionic substances on electronic equipment: amounts, sources, and effects. *Proceedings of the International Congress on Metallic Corrosion*, Toronto, Canada 3–7 June, pp. 296–303.
- Sinclair J. D., Psota-Kelty L. A. and Weschler C. J. (1985) Indoor/outdoor concentrations and indoor surface accumulations of ionic substances. *Atmospheric Environment* 19, 315–323.
- Sinclair J. D., Psota-Kelty L. A. and Weschler C. J. (1988a) Indoor/outdoor ratios and indoor surface accumulations of ionic substances at Newark, New Jersey. *Atmospheric Environment* 22, 461–469.
- Sinclair J. D., Psota-Kelty L. A., Weschler C. J. and Shields H. C. (1990) Deposition of airborne sulfate, nitrate and chloride salts as it relates to corrosion of electronics. *J. Electrochem. Soc.* 136, (in press).
- Sinclair J. D. and Weschler C. J. (1985) Estimating the indoor deposition rates of acidic substances from outdoor concentrations. *Proceedings of the Symposium on Corrosion Effects of Acid Deposition and Corrosion of Electronic Materials* (edited by Mansfeld F., Haagenrud S. E., Kucera V., Haynie F. H. and Sinclair J. D.) 168th Meeting of the Electrochemical Society, 13–18 October, Las Vegas, Nevada, pp. 258–266.
- Weschler C. J., Kelty S. P. and Lingousky J. E. (1983) The effect of building fan operation on indoor/outdoor dust relationships. *J. Air Pollut. Control Ass.* 33, 624–629.
- Weschler C. J. and Shields H. C. (1988) The influence of HVAC operation on the concentrations of indoor airborne particles. In *IAQ 88: Engineering Solutions to Indoor Air Problems*. American Society of Heating, Refrigerating and Air-Conditioning Engineers, Atlanta, 166–181.
- Weschler C. J. and Shields H. C. (1989) The effects of ventilation, filtration and outdoor air on the composition of indoor air at a telephone office building. *Envir. Int.* 15, 593–604.
- Weschler C. J., Shields H. C., Kelty S. P., Psota-Kelty L. A. and Sinclair J. D. (1989) Comparison of effects of ventilation, filtration, and outdoor air on indoor air at telephone office buildings: a case study. In *Design and Protocol for Monitoring Indoor Air Quality* (edited by Nagda N. L. and Harper J. P.) pp. 9–34. American Society for Testing and Materials, Philadelphia, P.A.
- Wesely M. L., Hicks B. B., Dannevik W. P., Frisella S. and Hausar R. B. (1977) An eddy-correlation measurement of particulate deposition from the atmosphere. *Atmospheric Environment* 11, 561–563.