

SUMMARY

S. Renes, B.P. Leaderer, L. Schaap, H. Verstraelen and T. Tosun. An Evaluation of Sink Terms in Removing NO₂ and SO₂ from Indoor Air. Efforts to model concentrations of indoor air contaminants require accurate information on the generation and removal mechanisms of the air contaminants. For reactive air contaminants the sink rate is an important removal mechanism. The sink or removal rate for two reactive indoor air contaminants (NO₂ and SO₂) were evaluated in an environmental chamber as a function of material type (painted sheetrock, wallpaper and carpeting), variable surface area of the material, relative humidity and air mixing. Sink rates for SO₂ are generally higher than those for NO₂. The sink rates for NO₂ and SO₂ were found to increase with material surface roughness and material surface area. Increases in relative humidity had a pronounced positive impact on SO₂ sink rates and a smaller but significant impact on NO₂ sink rates. The degree of air mixing was found to have a pronounced impact of both SO₂ and NO₂ sink rates.

RESUME

S. Renes, B.P. Leaderer, L. Schaap, H. Verstraelen et T. Tosun. Etude des taux d'élimination du NO₂ et du SO₂ de l'air des intérieurs par sédimentation. Pour établir des modèles de concentration de contaminants dans l'air des intérieurs, il est nécessaire d'obtenir des données précises sur les mécanismes de production et d'élimination des divers contaminants de l'air. Un mécanisme important pour l'élimination des contaminants réactifs est le taux de sédimentation. Les taux de sédimentation ou ceux d'élimination pour deux contaminants réactifs de l'air des intérieurs (NO₂ et SO₂) ont été mesurés dans une enceinte micro-climatique en fonction du matériau utilisé (plâtre en panneaux, papier peint, et tapis), de la grandeur de la surface, de l'humidité relative, et de l'agitation de l'air. Les taux de sédimentation du SO₂ sont, en général, plus élevés que ceux du NO₂. Les taux de sédimentation du NO₂ et du SO₂ augmentent avec la rugosité de la surface du matériau et de sa superficie. Une augmentation de l'humidité relative fait croître le taux de sédimentation du SO₂ d'une façon considérable, et l'effet sur le taux de sédimentation du NO₂, quoique moins prononcé, est sensible. Le degré d'agitation de l'air exerce un effet considérable sur les taux de sédimentation du SO₂ et du NO₂.

KURZFASSUNG

S. Renes, B.P. Leaderer, L. Schaap, H. Verstraelen und T. Tosun. Untersuchung der Sinkraten beim Entzug von NO₂ und SO₂ aus Zimmerluft. Die Beschreibung der Konzentration von Luftverunreinigungen in Innenräumen erfordert genaue Information über Erzeugungs- und Entfernungsmechanismen dieser Luftverschmutzungen. Für die Entfernung von chemisch aktiven Luftverunreinigungen spielt die Sinkrate eine wichtige Rolle. In einer Klimakammer wurde die Sinkrate bzw. Entfernungsrate für zwei chemisch aktive Bestandteile von Zimmerluftverschmutzung (SO₂ und NO₂) in Abhängigkeit von Wandmaterial (getünchte Gipsplatte, Tapete, Teppich) und Wandoberfläche, sowie von relativer Luftfeuchte und Luftvermischung untersucht. Die Sinkrate von SO₂ ist in allgemeinen grösser als die von NO₂. Mit zunehmender Oberflächenrauigkeit und -grösse wurde ein Ansteigen der Sinkraten von NO₂ und SO₂ festgestellt. Höhere relative Luftfeuchte hatte eine ausgeprägte Zunahme der SO₂ Sinkrate zur Folge und zeigte einen kleineren aber deutlichen Effekt in der NO₂ Sinkrate. Der Grad der Luftauführung erwies sich als wichtiger Parameter sowohl für die SO₂ als auch für die NO₂ Sinkraten.

AN EVALUATION OF SINK TERMS IN REMOVING
NO₂ AND SO₂ FROM INDOOR AIR

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Introduction

Air contaminant concentrations in the indoor environment are a function of several variables including: contaminant production rates; infiltration or ventilation rates; air mixing; sink rates; effectiveness of air cleaning devices; and outdoor concentrations. Efforts to model concentrations of indoor air contaminants require accurate information on each of the above variables. The importance of the sink terms in removing indoor contaminants and the parameters which determine its strength are poorly understood. A recent field study of two important indoor air contaminants (NO₂ from unvented combustion sources and SO₂ from unvented kerosene space heaters) has demonstrated the importance of the sink term in determining the indoor concentrations of these contaminants as well as the variability of the sink rates (1,2). In this study the sink rates were shown to be at least as important as infiltration in removing NO₂ and SO₂ and vary greatly within the same residence from time period to time period as well as between residences. The results of this field study pointed to a need to characterize the sink rates for NO₂ and SO₂. We conducted a series of chamber studies aimed at evaluating the major factors determining the sink or removal rate of NO₂ and SO₂ in residences: type and surface area of materials, relative humidity (RH), and air mixing.

Methods

Chamber

An all aluminum environmental chamber, 34m³ with a total wetted surface area of 185.2 m², was used for these experiments. The chamber possessed excellent temperature and humidity control, using modulated flow chilled water for cooling and modulated steam coils and electric heating elements for heating. A desiccant air dryer was used to reduce the moisture content of the incoming air, when necessary.

Air entered the chamber via a plenum beneath the floor, streamed upward through 13,900 perforations, and left the chamber via four return ducts in the ceiling. The design allowed a volume flow of up to 2,000 cfm (1000 ls⁻¹) with low linear velocity and very rapid mixing. The volume flow from the supply air duct (recirculation rate) could be varied from 400 to 2000 cfm (200-1000 ls⁻¹), which corresponds to 20-100 air changes per hour (ach). A variable percentage of the recirculation air could comprise fresh ventilation air. The ventilation rate could vary from 10 to 400 cfm (5 to 200 ls⁻¹), or 0.35 to 20 ach.

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Materials

The materials placed in the chamber included painted sheetrock, wallpaper and carpet. Eleven 1.98 by 1.22 m panels of sheetrock were painted with two coats of a semi gloss paint to provide a total of 22 surfaces (53 m²). Ten 1.98 by 1.22 m panels of sheetrock were wallpapered with a textured vinyl wallpaper to provide a total of 20 surfaces (48.3 m²). Six 1.98 by 1.22 m sheets of plywood were covered with a rayon medium pile carpet to provide a total of 12 surfaces (29 m²); the carpet was glued to the plywood.

Experiments

CO₂, NO₂ and SO₂ were injected into the chamber from bottled gas, and the decay curves recorded continuously by use of a Beckman model LB-2 infrared CO₂ analyzer, a Monitor Labs model 8440 chemiluminescence NO_x analyzer and a Monitor Labs model 8850 fluorescence SO₂ analyzer. The decay rates of the gases for two different surface areas of the materials and an empty chamber were recorded for a range of relative humidities under a constant ventilation rate of 2.2 ach and rapid mixing (recirculation rate of 20 ach or greater). Relative humidities were typically at 33%, 55% and 75% with temperature variations between 25°C and 37°C. In another set of experiments, the decays of CO₂, NO₂ and SO₂ were measured for two different surface areas of the materials under the conditions of no air recirculation and an air exchange rate of 0.35 ach. All experimental conditions were run in triplicate. After each run the chamber was flushed at a high ventilation rate for several hours.

The baseline concentration of the gases, C₀ (background levels), was recorded before each experiment and subtracted from all measurements. The gases were injected into the chamber until peak concentrations (C₁) of CO₂ = 1%, NO₂ = 1 ppm and SO₂ = 1 ppm were reached, typically over a two minute period. The decrease in the gas concentration in the chamber, under well mixed conditions, and upon cessation of gas injection, can then be represented by the following equation

$$C_t = C_1 e^{-kt} \quad (1)$$

where: C_t is the concentration of the gas (ppm) in the chamber at time t (hr); C₁ is the initial concentration of the gas (ppm) in the chamber after gas injection has stopped; k is the removal rate of the gas (hr⁻¹) due to ventilation and sink rates (i.e., absorption, chemical transformation, etc.); and t is the time in hours. The removal rate, k, in this analysis is assumed to follow a first order decay, given by:

$$k = n + m + RH \quad (2)$$

where: n is the infiltration rate (hr⁻¹); m is the absorption rate of the materials considered (hr⁻¹); and RH is the impact of relative humidity (hr⁻¹). Temperature variations encountered in these experiments were assumed to have a small or negligible effect. This simple model does not evaluate the potential for interactive terms, i.e., RH interaction with the various materials. The decays of the gases in ach or hr⁻¹ from peak concentrations were calculated from a semilog plot of concentration (minus background) vs time, using regression analysis. CO₂ decay rates were used to assess the outside air ventilation rate, n. The CO₂ decay rates for each experiment were subtracted from NO₂ and SO₂ decay rates to assess the impact of relative humidity and materials on the removal or sink rates (m and RH in equation 2).

Results and Discussion

For the same chamber ventilation rate settings, CO₂ decays were not found to vary as a function of relative humidity, temperature or surface area of the materials evaluated. Decay rates for NO₂ and SO₂ for an empty chamber and low relative humidity are assumed to be approximated by the CO₂ decay rate, since aluminum (the wetted surface of the chamber) is non-reactive. The impact of varying relative humidities on the sink rates for NO₂ and SO₂ under the conditions of a constant ventilation rate, rapid mixing and no materials in the chamber is shown in Figure 1. SO₂ sink rates demonstrated a pronounced increase with increasing RH, while a less pronounced variation in NO₂ sink rates under the same conditions was observed. Presumably the water solubility of the gases accounts for the sink rates, although potential wetting of the chamber surface area under higher RH may increase the efficiency of the surface in removing the gases. The impact of relative humidity on NO₂ sink rates observed in this study was less than reported by one other study (3).

The NO₂ and SO₂ sink rates versus % RH for variable surface areas of different materials under a constant ventilation rate, variable temperature and rapid mixing are shown in Figures 2 and 3. The sink rates for SO₂ (a range of 0.4 to 3.5 ach) were greater than NO₂ (a range of 0.05 to 1.5 ach) for all conditions, representing the greater reactivity of SO₂. A pronounced effect of material type, material surface area and RH was observed for SO₂ (Fig. 2). The NO₂ sink rates (Fig. 3) were found to increase with increasing RH and material surface area for wall paper and carpet. The impact of RH and variable surface area for painted board on NO₂ sink rates was less pronounced but above the ventilation rates measured by CO₂ decays. Temperature generally did not correlate with the sink rates for NO₂ over the range of temperatures encountered in these experiments. SO₂ sink rates demonstrated a significant but low negative correlation with temperature, reflecting the fact that higher relative humidities in these experiments for the more water soluble SO₂ were achieved under conditions of lower temperature. There was no evidence of NO₂ conversion to NO or re-emission of NO₂ from the materials tested.

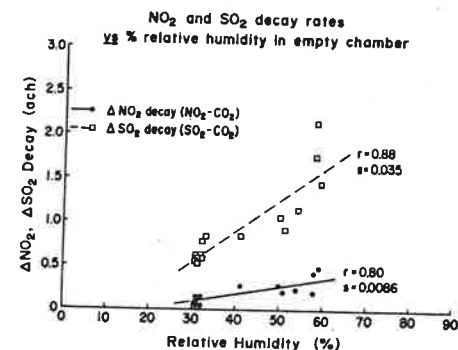


Fig. 1. The sink or removal rates in air changes per hour (ach or hr⁻¹) for NO₂ and SO₂ as a function of percent relative humidity (% RH) in the empty all aluminum environmental chamber.

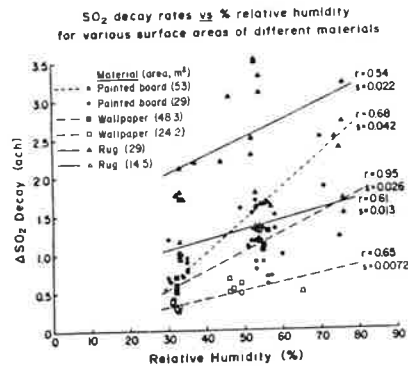


Fig. 2. The sink or removal rates in air changes per hour (ach or hr⁻¹) for SO₂ as a function of percent relative humidity (% RH) for two surface areas of three materials.

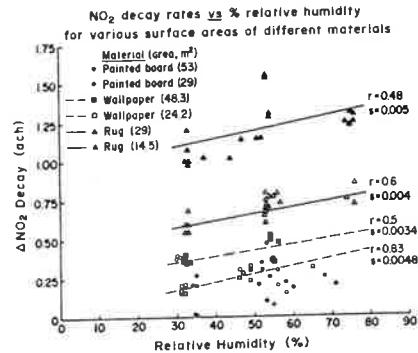


Fig. 3. The sink or removal rates in air changes per hour (ach or hr⁻¹) for NO₂ as a function of percent relative humidity (% RH) for two surface areas of three materials.

NO₂ and SO₂ sink rates were found to vary as a function of mixing. The NO₂ and SO₂ sink rates for variable surface area of the materials and empty chamber, while controlling for RH, were generally a factor of 2 to 3 lower for the condition of the air recirculation rate of the chamber set equal to zero. However, we have no way of knowing what the mixing rates were under these conditions. Further study of the impact of air mixing on the sink rates is needed.

The data presented in Figures 1, 2 and 3 can be combined and analyzed using multivariate techniques (regression) to develop a model to predict the sink rates for NO₂ and SO₂ as a function of relative humidity, type of material and total surface area of the material. The analysis assumes a linear relationship and rapid mixing. This analysis does not examine interaction between relative humidity and the materials. The results of the analysis are shown in Table 1. In conditions where there is less than complete rapid mixing, the coefficients would be reduced considerably. Table 1 reflects the results presented in Figures 1, 2 and 3.

Conclusion

Sink rates are important in modeling indoor concentrations of NO₂ and SO₂. Sink rates for SO₂ are generally higher than those for NO₂. The sink rates for NO₂ and SO₂ were found to increase with material surface roughness and material surface area. Increases in relative humidity had a pronounced positive impact on SO₂ sink rates and a smaller but significant impact on NO₂ sink rates. The degree of air mixing is an important variable in determining the sink rates for NO₂ and SO₂. These experiments evaluated three types of materials found in residences; evaluations of other materials commonly found in residences (e.g., draperies, natural vs man made fibers, other types of carpets, etc.) are needed.

Table 1

Regression Analysis[†] for Predicting the Impact of Relative Humidity and Variable Surface Areas of Materials on NO₂ and SO₂ Sink Rates^{††} in Air Changes per Hour (hr⁻¹) for Chamber Experiments Under Conditions of Rapid Mixing

Parameters	Sink Rate of NO ₂ (hr ⁻¹)	Sink Rate of SO ₂ (hr ⁻¹)
<u>Intercept</u>	0.043 (0.108)	-0.094 (0.432)
<u>Coefficients</u>		
1) Surface area of carpet (m ²)	0.033 (0.0013)	0.054 (0.0051)
2) Surface area of wallpaper (m ²)	0.0039 (0.0009)	0.0018 (0.0035)
3) Surface area of painted board (m ²)	0.001 (0.0007)	0.0101 (0.0029)
4) Relative Humidity (%)	0.0033 (0.0009)	0.0173 (0.0034)
<u>R²</u>	0.92	0.70
Degrees of Freedom	4, 94	4, 94

[†] the standard error of the estimate is presented in parenthesis

^{††} m and RH in equation 2.

The results of these chamber experiments indicate that field studies directed toward assessing and modeling exposures to indoor air contaminants should include information on the interior surface areas, furnishings, air mixing and relative humidities in the residences studied, in order to determine the sink rates for reactive air contaminants. Failure to gather information needed to estimate the sink rates for reactive air contaminants can result in large errors in predicted indoor contaminant levels.

Acknowledgements

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References

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