

# Nitrous acid concentrations in homes and offices in residential areas in Greater Cairo

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Received 19th February 2002, Accepted 1st May 2002

First published as an Advance Article on the web 24th May 2002

Indoor and outdoor measurements of nitrous acid and nitrogen dioxide were conducted at four homes and two offices in residential areas in Greater Cairo during winter (2000–2001) and summer (2001) seasons. Indoor nitrogen dioxide concentrations were higher than outdoor levels at the four homes, whereas indoor concentrations of nitrogen dioxide were lower than outdoor levels at the two offices, during both seasons. Indoor nitrous acid concentrations were higher than outdoor levels at all homes and offices during the period of study. The mean indoor nitrous acid concentrations were 6.8 ppb and 3.67 ppb in the four homes, whereas they were 1.42 ppb and 1.24 ppb in the two offices, during the winter and summer seasons, respectively. Indoor/outdoor ratios of nitrous acid concentration were 6.94 in the winter and 5.03 in the summer for all of the homes. However, the ratios were 1.31 and 1.61 during the winter and summer seasons, respectively, for the two offices. Insignificant positive correlation coefficients were found between indoor and outdoor concentrations of nitrous acid at homes and offices. The maximum outdoor nitrous acid concentrations were recorded during the winter season. Significant positive correlation coefficients were found between nitrous acid and nitrogen dioxide and relative humidity in homes and offices. The ratios of nitrous acid to nitrogen dioxide concentrations ranged from 0.045 to 0.16, with a mean of 0.1, in the four homes, whereas the ratios ranged from 0.026 to 0.09, with a mean of 0.059, in the two offices.

## Introduction

Nitrogen dioxide levels in the ambient atmosphere are closely related to the location of major sources, meteorological factors and atmospheric conversion reactions.<sup>1–3</sup> Nitrogen dioxide is produced indoors from unvented or poorly vented combustion devices and by tobacco smoking.<sup>4–7</sup> Indoor nitrogen dioxide levels are dependent on indoor and outdoor sources, air infiltration and ventilation, chemical reactions and adsorption or absorption of the gas on indoor surfaces.<sup>8–11</sup>

Nitrous acid (HONO) is formed in the gas phase during combustion and by the heterogeneous hydrolysis of nitrogen dioxide.<sup>12–14</sup> The indoor production of HONO has been described previously in scenarios of elevated NO<sub>2</sub> concentrations produced by injection of NO<sub>2</sub> into the indoor space<sup>13</sup> or by unvented indoor gas combustion appliances.<sup>15,16</sup> Nitrous acid may be found in higher concentrations indoors as a result of surface catalysed reactions involving NO<sub>2</sub> and water.<sup>12,17,18</sup> Spicer *et al.*<sup>19</sup> demonstrated that gas ranges and heaters are sources of HONO(g) emissions. Measurements in research houses and in a mobile laboratory showed that elevated indoor HONO concentrations resulted from unvented indoor combustion.<sup>10,16,20</sup> Nitrous acid is less reactive and persists in indoor air for an extended period of time.<sup>21,22</sup> Peak HONO concentrations of 30–50 ppb were found in a mobile office-home when a conventional gas stove produced NO<sub>2</sub> concentrations of 0.3–1 ppm.<sup>15,16</sup> The average indoor concentrations of HONO in homes were reported to be 5.44 ppb and 2.27 ppb during the winter and summer seasons, respectively.<sup>23</sup> Spengler *et al.*<sup>22</sup> found that the mean concentration of HONO was 4.7 ppb in homes in Albuquerque, New Mexico. In 47 residences in state college, Pennsylvania, Suh *et al.*<sup>24</sup> found that the level of HONO in homes with gas stoves averaged 3.1 ppb, whereas it was 1.1 ppb in homes without gas stoves. Maximum HONO concentrations in the range 5–10 ppb were measured in four different home environments with a small kerosene heater in operation.<sup>25</sup>

Nitrous acid plays an important role in tropospheric

chemistry. The sunlight photolysis of HONO may be the major source of OH radicals that promote ozone formation.<sup>26</sup> Nitrogen dioxide is a precursor of HONO in both urban and rural environments and its formation is governed by heterogeneous reactions.<sup>27</sup> Moreover, significant levels of HONO have been detected in car exhausts.<sup>28,29</sup> Rondon and Sanhueza<sup>30</sup> also recorded high levels of HONO during biomass burning in tropical savannah. The nitrous acid concentration was low during the daytime and accumulated through the night.<sup>31</sup> Elevated concentrations of nitrous acid, up to 3.2 ppb, were detected in the suburban area of Zurich, Switzerland, and the correlation with NO<sub>x</sub> and black carbon suggests a heterogeneous formation of HONO.<sup>32</sup> Mean HONO concentrations in tunnel exhaust and background air were 6.9 ppb and 0.7 ppb, respectively.<sup>33</sup>

Nitrous acid may adversely affect human health: it reacts rapidly with gaseous secondary amines in air to form carcinogenic nitrosamines.<sup>34–36</sup> This raises the possibility of the *in vivo* formation of nitrosamines from the inhalation of gaseous HONO.<sup>37,38</sup> Short-term exposures to elevated concentrations of nitrous acid may be irritating to mucous membranes in healthy subjects and may alter lung function in asthmatic subjects.<sup>39,40</sup>

There is a lack of information on HONO concentrations in urban areas of subtropical semiarid developing countries under the prevailing air quality and climate conditions. Cairo has a desert climate characterized by very dry heat. The monthly mean temperatures range from 14 °C in January to 29 °C in July. The maximum daily temperature in July can reach 43 °C. The average relative humidity is 56%. Nights are generally cool and, during the winter, quite damp; radiational cooling often leads to shallow, stable inversions.

This study aims to evaluate the indoor HONO concentrations in homes and offices in Greater Cairo, and to compare these concentrations with ambient air levels. In addition, factors that influence indoor HONO concentrations are considered.

## Materials and methods

Indoor and outdoor measurements of HONO and NO<sub>2</sub> concentrations were made at four homes and two offices in suburban residential areas in Greater Cairo. The areas of the homes varied from 65 to 120 m<sup>2</sup>. Each home was occupied by a single family. One home was occupied by a light smoker. All homes used gas appliances for cooking and electricity for water heating. Indoor air samples in homes were collected from the living room. The two offices had areas of 20 and 24 m<sup>2</sup>. Neither of the two offices had indoor sources of NO<sub>2</sub>. All homes and offices had natural ventilation (windows and doors). Because of the warm winter and mild summer, most of Cairo's homes and offices have no heating systems. The indoor samples were collected at approximately 1.5 m above the floor and in the centre of the room. Outdoor samples were collected at the same time as indoor samples. The samples were taken daily (every 24 h) during the winter (December 2000–February 2001) and summer (June–August 2001) seasons.

## Chemical analysis

The sodium carbonate-coated denuder sampling technique was used for the measurement of indoor and outdoor HONO, as described in detail by Ferm and Sjodin.<sup>41</sup> Samples were collected using two denuders in series. The denuders were mounted vertically inside a box to avoid particle settling on the denuder walls due to gravity. Air was drawn at 2 L min<sup>-1</sup> through the two denuders using a calibrated pump, which yields a collection efficiency for HONO of approximately 96% in each denuder.<sup>41</sup> After each sampling time, the denuders were sealed and taken to the laboratory. The denuders were leached separately with distilled water, and the nitrite contents in each leaching solution were analysed spectrophotometrically. Two denuders in series were used to account for interferences in sampling HONO. Nitrous acid concentrations were calculated according to the equation:<sup>41</sup>

$$\text{HNO}_2 = (a - b) \times 1.085$$

where *a* and *b* represent the nitrite contents per volume of sampled air in the first and second denuder in the series, respectively. Nitrite collection in the second denuder was typically around 16% of that in the first. It was calculated that an average of 1.5% of the ambient NO<sub>2</sub> was captured and converted to nitrite in the sodium carbonate layer. The detection limit for HONO was 0.01 ppb for 24 h sampling.

Nitrogen dioxide samples were collected in glass bubblers containing sodium hydroxide–sodium arsenite solution as an

absorbing reagent using a pump calibrated to draw 0.5 L min<sup>-1</sup>. The absorbent was then reacted with phosphoric acid, sulfanilamide and *N*-1-(naphthyl)ethylenediamine dihydrochloride to form an azo dye.<sup>42</sup> The absorbance of the sample against the reagent blank was measured at 540 nm by a spectrophotometer, and the NO<sub>2</sub> concentration was calculated from the calibration standard curve and volume of air.

The indoor relative humidity was measured during every sampling using a Sigma-II thermohygrograph (No. 7210, SK Sato Keiryoki MFG-Co., Ltd., Japan). The instrument was calibrated at the factory and is regularly calibrated at the Egyptian meteorological authority.

## Statistical analysis

The correlation coefficient (*r*) and the correlation significant *t*-test were determined using the alternative method of calculation.<sup>43</sup> Student's *t*-test was used to estimate the significant difference between the mean concentrations during the winter and summer seasons and between indoor and outdoor concentrations.<sup>43</sup>

## Results and discussion

The indoor concentrations of nitrogen dioxide (NO<sub>2</sub>) inside the four homes are shown in Table 1. The highest levels of NO<sub>2</sub> were recorded inside home 1, and significant differences (*P* < 0.05) were found between the average concentrations of NO<sub>2</sub> in homes 1 and 3 and in homes 1 and 4, possibly due to indoor smoking. The indoor NO<sub>2</sub> concentrations in the four homes were higher than the outdoor levels during the winter and summer seasons (Table 2), and the differences were statistically significant (*P* < 0.001). The indoor/outdoor ratios of the NO<sub>2</sub> concentrations for all homes were in the range 1.20–2.34 with a mean value of 1.76 ± 0.298 during the winter, and in the range 1–1.91 with a mean value of 1.34 ± 0.212 during the summer (Fig. 1). Increased indoor NO<sub>2</sub> concentrations have previously been associated with homes using gas cooking.<sup>5,6</sup> Indoor levels of NO<sub>2</sub> were higher than outdoor concentrations in homes using gas cooking.<sup>11,44,45</sup> In the present study, the mean indoor concentrations of NO<sub>2</sub> were 65 ppb and 39 ppb during the winter and summer seasons, respectively, in the four homes (Table 2), and the difference was statistically significant (*P* < 0.001). The higher indoor concentration of NO<sub>2</sub> during the winter season may be due to an increased use of gas cooking appliances and a lower natural ventilation, as windows and doors are closed for long periods, leading to the accumulation of NO<sub>2</sub> in indoor air. The highest

**Table 1** Nitrogen dioxide and nitrous acid concentrations (ppb) inside the different investigated homes and offices during the winter and summer seasons (2000–2001)

Site	Size/m <sup>2</sup>	Pollutant	Winter		Summer		Average	
			Range	Mean	Range	Mean	Range	Mean
<b>Homes<sup>a</sup></b>								
Home 1 <sup>b</sup>	85	NO <sub>2</sub>	48–120	80.8	30–73	49.7	30–120	65.3
		HONO	3.89–12.5	8.65	2.4–7.3	4.65	2.4–12.5	6.65
Home 2	65	NO <sub>2</sub>	33–104	66.9	21–61	39.1	21–104	53
		HONO	2.94–11.22	7.2	1.6–6.5	3.5	1.6–11.22	5.35
Home 3	120	NO <sub>2</sub>	27–81	54	20–48	31.5	20–81	42.5
		HONO	2.1–9.4	5.6	1.3–5.7	3.12	1.3–9.4	4.36
Home 4	100	NO <sub>2</sub>	29–92	58.1	23–56	35.5	23–92	46.8
		HONO	1.6–9.9	5.84	1.3–6.2	3.31	1.3–9.9	4.57
<b>Offices<sup>a</sup></b>								
Office 1	24	NO <sub>2</sub>	14–28	22	15–35	25.7	14–35	23.85
		HONO	0.87–2.21	1.75	0.52–2.13	1.42	0.52–2.21	1.59
Office 2	20	NO <sub>2</sub>	13–25	19.5	16–29	22.2	13–29	20.85
		HONO	0.6–1.67	1.10	0.53–1.71	1.05	0.53–1.71	1.08

<sup>a</sup>All homes and offices have natural ventilation and do not use any type of heating system. <sup>b</sup>Occupied by a light smoker.

**Table 2** Indoor and outdoor concentrations of nitrous acid and nitrogen dioxide (ppb) at the four homes during the winter and summer seasons (2000–2001)<sup>a</sup>

Parameter	Location	Winter				Summer			
		N	Range	Mean	S.D.	N	Range	Mean	S.D.
NO <sub>2</sub>	In	40	27–120	65	25.49	40	20–73	39	14.2
	Out	40	20–55	37	9.19	40	16–44	29	7.3
HONO	In	40	1.6–12.5	6.8	2.97	40	1.3–7.3	3.67	1.51
	Out	40	0.41–1.9	0.98	0.41	40	0.32–1.3	0.73	0.27

<sup>a</sup>In, indoor; Out, outdoor; N, number of samples; S.D., standard deviation.

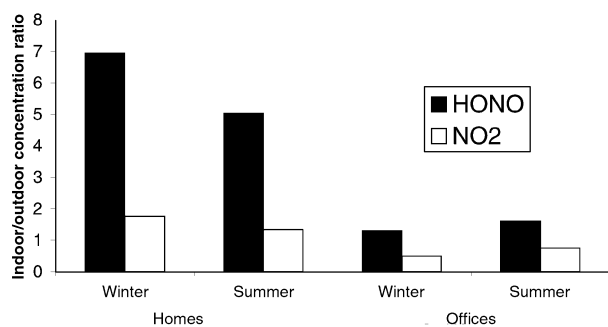
indoor NO<sub>2</sub> concentrations were also observed during the winter season in other studies.<sup>11,46</sup>

In the present study, the highest levels of nitrous acid (HONO) were recorded in home 1 (Table 1), and significant differences ( $P < 0.05$ ) were found between the average concentrations of HONO in homes 1 and 3 and in homes 1 and 4, due to the higher NO<sub>2</sub> concentrations in home 1 from both gas cooking appliances and smoking. Based on the indoor and outdoor data at the four homes (Table 2), the indoor concentrations of HONO were higher than the outdoor levels during the winter and summer seasons, and the differences were statistically significant ( $P < 0.001$ ). The indoor/outdoor ratios of the HONO concentration were in the range 1.78–16.32 with a mean value of  $6.94 \pm 3.47$  in the winter, and in the range 1.45–10.33 with a mean value of  $5.03 \pm 2.15$  in the summer, for all of the homes (Fig. 1). In addition, insignificant positive correlation coefficients were found between the indoor and outdoor concentrations of HONO: 0.30 in the winter and 0.32 in the summer. These results indicate that HONO is formed inside homes from gas combustion, and therefore may be produced from both heterogeneous reactions involving NO<sub>2</sub> and primary production during combustion.<sup>10</sup> Nitrous acid levels in homes have been found to be higher than those outdoors, with the highest concentrations inside homes with gas cooking stoves.<sup>22,23</sup> In the present study, the mean indoor concentrations of HONO were 6.8 ppb and 3.67 ppb during the winter and summer seasons, respectively, in the four homes (Table 2), and the difference was statistically significant ( $P <$

0.001). The higher concentrations of HONO during the winter season may be attributed to the accumulation of both HONO and its precursor (NO<sub>2</sub>) in indoor air due to lower natural ventilation. This result is in agreement with the observation of Brauer *et al.*,<sup>23</sup> who found that the highest indoor concentration of HONO was found during the winter and the lowest concentration was recorded during the summer in homes. In our study, the mean concentrations of HONO inside the four homes (Table 2) were lower than those reported by Biermann *et al.*<sup>15</sup> and Pitts *et al.*<sup>16</sup> In contrast, the mean indoor concentration of HONO during the winter season in the present study was higher than that reported in comparative studies.<sup>22–24</sup>

The average indoor concentrations of NO<sub>2</sub> were 23.85 ppb in office 1 and 20.85 ppb in office 2 (Table 1), and the difference was not statistically significant. At the two offices (Table 3), the indoor levels of NO<sub>2</sub> were lower than the outdoor concentrations during the winter and summer seasons, and the differences were statistically significant ( $P < 0.01$ ). The indoor/outdoor ratios of the NO<sub>2</sub> concentration for the two offices were in the range 0.41–0.54 with a mean of  $0.5 \pm 0.027$  during the winter, and in the range 0.59–0.87 with a mean of  $0.75 \pm 0.057$  during the summer (Fig. 1). Outdoor NO<sub>2</sub> is the main source of indoor NO<sub>2</sub> in the offices. The mean indoor concentrations of NO<sub>2</sub> were 21 ppb in the winter and 24 ppb in the summer at the two offices (Table 3), and the difference was not statistically significant. These results are in agreement with those of Muramatsu<sup>47</sup> and Hassan,<sup>11</sup> who found that NO<sub>2</sub> concentrations inside offices were lower than outdoor values. Indoor levels of NO<sub>2</sub> are less than outdoor concentrations in microenvironments without combustion sources.<sup>4</sup>

Although the indoor concentrations of NO<sub>2</sub> in the two offices were lower than the outdoor levels in this study, the indoor HONO concentrations were greater than the outdoor values during both seasons (Table 3), and the differences were statistically significant ( $P < 0.05$ ). The indoor/outdoor ratios of the HONO concentration at the two offices were  $1.31 \pm 0.80$  (range, 0.48–4.12) and  $1.61 \pm 0.71$  (range, 0.58–3.1) during the winter and summer seasons, respectively (Fig. 1). In addition, insignificant positive correlation coefficients were found between the indoor and outdoor concentrations of HONO: 0.32 in the winter and 0.34 in the summer. This is in agreement with the observation of Lee *et al.*,<sup>48</sup> who found that the indoor and outdoor concentrations of HONO in an empty lecture room were not statistically correlated. Nitrous acid does not



**Fig. 1** Indoor/outdoor concentration ratios for nitrous acid and nitrogen dioxide at all homes and offices during the winter and summer seasons.

**Table 3** Indoor and outdoor concentrations of nitrous acid and nitrogen dioxide (ppb) at the two offices during the winter and summer seasons (2000–2001)<sup>a</sup>

Parameter	Location	Winter				Summer			
		N	Range	Mean	S.D.	N	Range	Mean	S.D.
NO <sub>2</sub>	In	20	13–28	21	4.9	20	15–35	24	6.02
	Out	20	26–56	42	9.9	20	20–47	32	8.5
HONO	In	20	0.61–2.21	1.42	0.52	20	0.52–2.13	1.24	0.49
	Out	20	0.45–2.01	1.08	0.41	20	0.38–1.51	0.77	0.31

<sup>a</sup>In, indoor; Out, outdoor; N, number of samples; S.D., standard deviation.

accumulate in ambient air and is rapidly photolysed during the day.<sup>49</sup> In the present study, the increased concentrations of HONO inside the offices (Table 3) may be due to the heterogeneous conversion of NO<sub>2</sub> that has infiltrated from outdoors. Consistent with the observations of previous investigators, indoor HONO production results from the reactions of NO<sub>x</sub> species which have penetrated indoors from outdoor air.<sup>21,48</sup> One possible mechanism for indoor HONO production is the heterogeneous reaction of NO<sub>2</sub> with water.<sup>18</sup> Also, in a study conducted by Pitts *et al.*,<sup>13</sup> NO<sub>2</sub> was injected into the air of a mobile laboratory, and HONO concentrations increased with first-order kinetics with respect to NO<sub>2</sub>. In the present study, HONO concentrations in the homes were higher than those found in the offices (Table 1), due to the use of gas cooking appliances in the homes. The mean concentrations of HONO inside the two offices were 1.42 ppb and 1.24 ppb during the winter and summer seasons, respectively (Table 3), and the difference was not statistically significant. These results are higher than those reported by Poon *et al.*,<sup>50</sup> who found that the concentration of HONO inside a clean room ranged from 0.10 ppb in the summer to 0.61 ppb in the winter. However, the HONO levels in the present study were within the range of the result observed by Lee *et al.*,<sup>48</sup> who found that the mean indoor concentration of HONO was 1.1 ppb, with an indoor/outdoor ratio of 1.6, during the summer season in an empty lecture room.

Significant positive correlation coefficients ( $P < 0.001$ ) were found between the HONO and NO<sub>2</sub> concentrations and between the HONO concentration and relative humidity inside the homes and offices during both seasons (Figs. 2–5). This suggests that the indoor production of HONO increases with increasing NO<sub>2</sub> concentration and relative humidity, and may result from the reaction of NO<sub>2</sub> and H<sub>2</sub>O on indoor surfaces. Indoor concentrations of HONO increase with increasing indoor NO<sub>2</sub> concentrations and relative humidity.<sup>22,23,51</sup> In the present study, the ratios of HONO to NO<sub>2</sub> were in the range 0.045–0.16, with a mean of  $0.10 \pm 0.08$ , in the four homes. However, the ratios ranged from 0.026 to 0.09, with a mean of  $0.059 \pm 0.015$ , in the two offices; these values were much lower than those found in the homes. The higher HONO to NO<sub>2</sub> concentration ratios found in the homes is probably due to the higher production of HONO as a result of the rapid

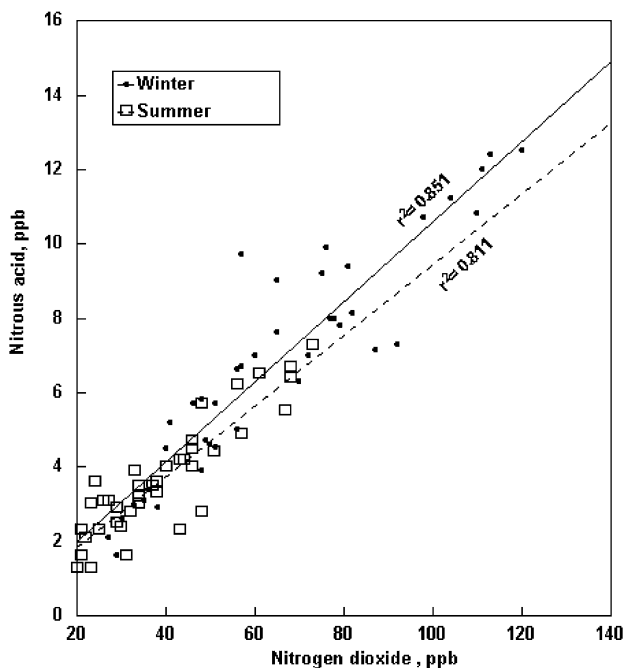


Fig. 2 Scatter diagram of nitrogen dioxide and nitrous acid concentrations inside the four homes.

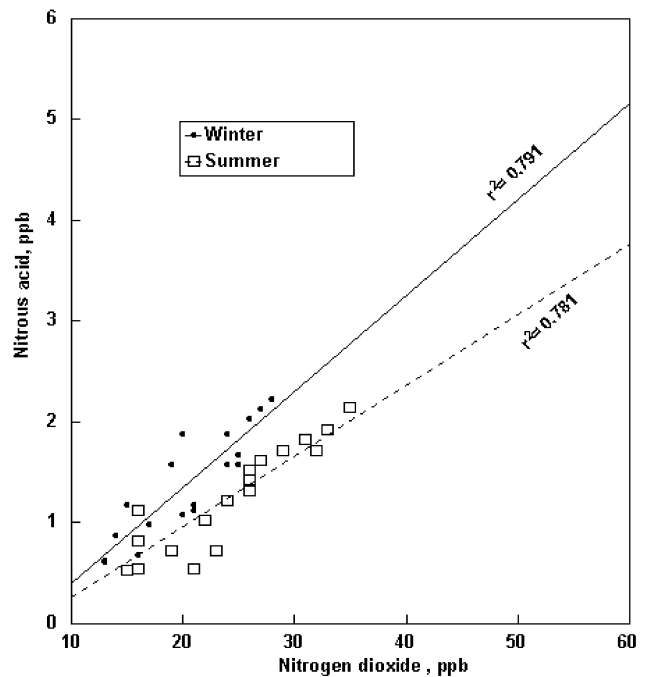


Fig. 3 Scatter diagram of nitrogen dioxide and nitrous acid concentrations inside the two offices.

build-up of HONO and NO<sub>2</sub> during gas combustion. Pitts *et al.*<sup>16</sup> found that the HONO production rate was higher when NO<sub>2</sub> was generated by combustion than when pure NO<sub>2</sub> was injected into a mobile laboratory, and the elevated HONO concentrations found during gas combustion may be associated with direct emissions of HONO from the combustion flame, in addition to heterogeneous processes. Pitts *et al.*<sup>16</sup> found that the ratios of HONO to NO<sub>2</sub> ranged from 0.15 to 0.30 in all of the indoor experiments with the gas stoves, whereas the ratio was  $\sim 0.04$  when pure NO<sub>2</sub> was injected into a mobile laboratory. In a series of controlled gas range emission tests in research homes, HONO to NO<sub>2</sub> ratios at peak NO<sub>2</sub> concentrations were 0.08–0.15.<sup>10</sup> Spengler *et al.*<sup>22</sup> measured HONO in homes located in Albuquerque, New Mexico, and

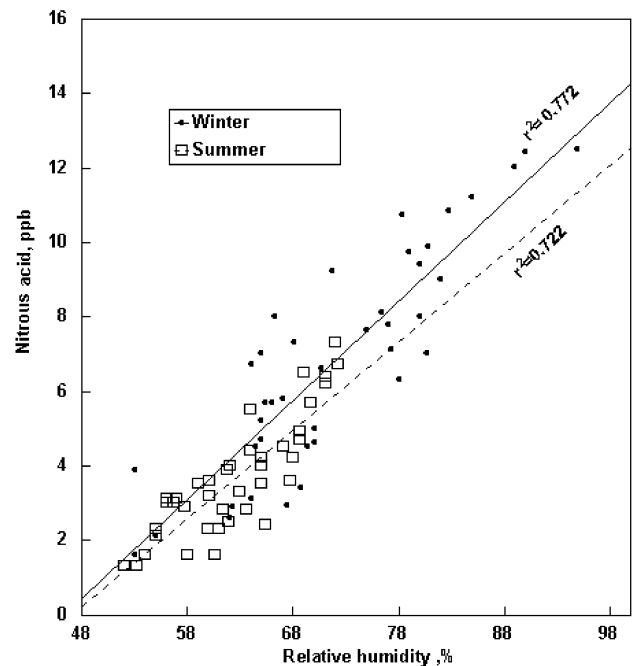


Fig. 4 Scatter diagram of relative humidity and nitrous acid concentrations inside the four homes.

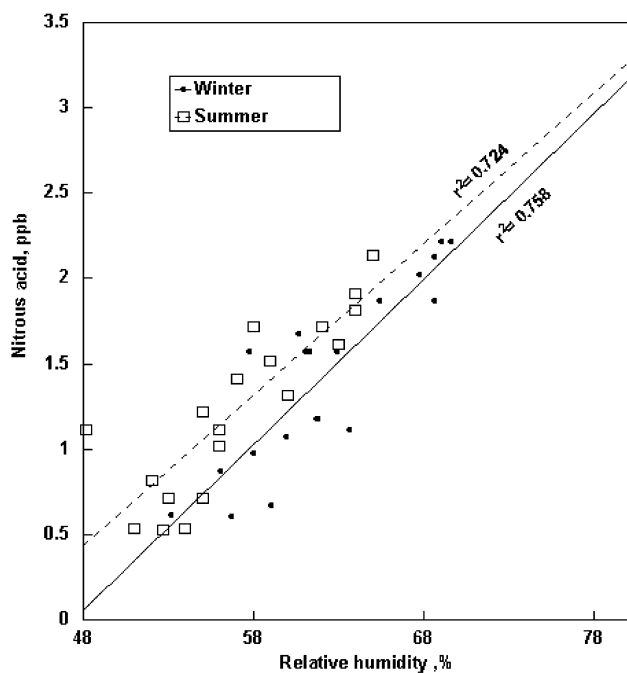


Fig. 5 Scatter diagram of relative humidity and nitrous acid concentrations inside the two offices.

found HONO concentrations ranging from 5% to 15% of the measured indoor NO<sub>2</sub> concentrations. In the present study, inside the homes, significant positive correlation coefficients ( $P < 0.01$ ) were found between the indoor/outdoor ratio of the HONO concentration and NO<sub>2</sub> ( $r = 0.57$  during the winter and  $r = 0.63$  during the summer), as well as between the indoor/outdoor ratio and relative humidity ( $r = 0.51$  in the winter and  $r = 0.54$  in the summer). However, weak positive correlation coefficients were found between the HONO/NO<sub>2</sub> ratio and NO<sub>2</sub> concentration ( $r = 0.2$ – $0.45$ ) and between the HONO/NO<sub>2</sub> ratio and relative humidity ( $r = 0.29$ – $0.46$ ) in homes and offices, as well as between the indoor/outdoor ratio of the HONO and NO<sub>2</sub> concentration ( $r = 0.44$ – $0.46$ ) and between the indoor/outdoor ratio and relative humidity ( $r = 0.31$ – $0.47$ ) in the offices during both seasons.

Nitrous acid is emitted in motor vehicle exhausts and is also formed in the atmosphere from the conversion of nitrogen oxides.<sup>33</sup> The concentrations of HONO exhibit a diurnal variation with maximum levels at night due to daytime photolysis.<sup>49</sup> In the present study, the mean outdoor concentrations of HONO were 0.98 ppb and 1.08 ppb during the winter, and 0.73 ppb and 0.77 ppb during the summer, in homes and offices, respectively (Tables 2 and 3). The observed differences in values during the winter and summer seasons were statistically significant ( $P < 0.05$ ). The higher concentrations of HONO during the winter may be attributed to the effect of meteorological factors. The higher inversion during the winter leads to increased accumulation of both HONO and its precursors (NO and NO<sub>2</sub>). The rate and extent of the daytime photolysis of HONO are strongly reduced during the winter, due to the shorter time period between sunrise and sunset and the lower sunlight intensity; this preserves the high HONO concentrations through the day.<sup>52</sup> The highest HONO concentrations were observed during the winter season.<sup>53</sup>

## Conclusions

Indoor HONO concentrations were higher than outdoor levels, even when indoor NO<sub>2</sub> concentrations were lower than outdoor levels. Nitrous acid concentrations in homes were higher than those found in offices. This may be attributed to an increase in the concentration of both HONO and its precursor

(NO<sub>2</sub>) during gas combustion. The indoor/outdoor concentration ratios suggest that HONO is produced indoors. The indoor HONO concentration increases with increasing indoor NO<sub>2</sub> and relative humidity. This indicates that HONO is formed from heterogeneous reactions of NO<sub>2</sub> and H<sub>2</sub>O. The higher HONO to NO<sub>2</sub> concentration ratio found in homes is probably due to the higher production of HONO as a result of the rapid build-up of HONO and NO<sub>2</sub> during gas combustion.

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