Front-door Concentrations and Personal Exposures of Danish Children to Nitrogen Dioxide

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The aims of the study were to evaluate the front-door concentration of traffic exhaust fumes as a surrogate for the personal exposure of children and to study factors in the behavior and the environment of children that affect their personal exposure to nitrogen dioxide (NO₂). The exposure to NO₂ of 103 children living in Copenhagen and 101 children living in rural areas of Denmark was studied by measuring average concentrations over 1 week with diffusive badge samplers placed outside the front door of the home, inside the child's bedroom, and on each child. Detailed information about the activities of the children involving potential exposure to NO₂ was noted in diaries. The results indicated that the front-door concentration of traffic pollution might be used to classify the personal exposure of urban children, although misclassification would be introduced. Multiple regression analysis showed several factors that affected the personal NO₂ exposure of the children independently, including the front-door concentration, the bedroom concentration, time spent outdoors, gas appliances used at home, passive smoking, and burning candles. Key words: air pollution, children, exposure, nitrogen dioxide, traffic.

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Many epidemiological studies deal with the effects of air pollution on children's health (1–17). Some have focused specifically on traffic exhaust fumes and used the residential outdoor concentration or the residential traffic density as surrogates for personal exposure (15–17).

The principal aim of this study was to determine if the front-door concentration of traffic exhaust fumes can be used to classify the actual personal exposure of children. Ideally, this requires measurements of a marker exclusively related to traffic exhaust fumes. Such a marker has not, to our knowledge, yet been identified. Thus, we used NO₂ as a marker of exposure to traffic exhaust fumes; the design of the study ensured that the contributions from sources other than traffic were reduced substantially. Moreover, the data allowed us to study factors in the environment and the behavior of Danish children that affect their exposure to NO₂.

Methods

Selection of children. Maps and traffic counts were used to identify 109 streets in central Copenhagen, Denmark, with or near to high traffic density and 215 streets in rural areas 20–50 km outside Copenhagen with low traffic density and no nearby major source of NO₂. We used the Central Population Registry, in which information including the sex, age, name, and residential address is registered for the entire Danish population, to identify all children between 4 and 12 years of age who lived on the 324 streets. A procedure to ensure geographical variation within the two residential areas was used, and the families of the children were chosen at random. The families received an invitation by mail to participate in the study, and a short questionnaire covering resident smokers, gas appliances in the kitchen, and potentially polluting heating sources such as coke ovens, wood-burning stoves, and kerosene heaters. Participants were chosen on the basis of the low presence of these indoor sources of NO₂ in order to make the outdoor contribution dominant. Moreover, as Copenhagen is a city with few industries and a well-developed district heating system, traffic is the major source of outdoor NO₂ pollution (18). The rural areas are considered as reference in terms of traffic pollution.

NO₂ measurements. The measurements were carried out during 2 weeks in October 1994, 2 weeks in April 1995, 2 weeks in May 1995, and 1 week in June 1995. During each week, passive NO₂ samplers (badges) were placed in three locations each at approximately 15 urban dwellings and 15 rural dwellings: outside the front door, in the bedroom of the child, and on the child. The front-door badges were fixed under a cap of stainless steel. In the urban areas, they were typically placed 0.5 m from the fronts of the buildings, 4 m above street level, and within 10 m of the front door. In the rural areas, they were placed either on spears in gardens or 0.5 m from the fronts of the houses, 1.5 m above the ground, and within 10 m of the front door. In the bedrooms of the children, the samplers were placed 1–1.5 m above the floor and distant from the door, the window, and any source of heat. The children carried their personal badges outside their clothes, usually on a belt. When the children were bathing or doing sports, the badges were placed as close to them as possible; at night, the badges were placed beside the bed with the surface side up. Eight trained persons operating two by two started the measurements and gave the families careful instructions during one weekend and collected the badges the next weekend, sealed them and stored them in a freezer until analysis.

Diary notes. Each day the family filled in a printed diary covering the activities of the child; exact locations; time spent indoors, outdoors, in a car, in a bus, or in a tractor; time exposed to passive smoking; time when gas appliances were used in the kitchen at home; time spent near fire (for example burning candles, fireplaces, woodburning stoves, and barbecues); and time exposed to perceptible air pollution from point sources like factories. On the basis of the diary notes, we added the variable time spent in a city. For children in Copenhagen, a city was defined as within 10 km of the center of Copenhagen; for the rural children, suburbs of Copenhagen and towns with more than 20,000 inhabitants were also counted as cities. The variable time spent in a car or a bus was calculated as the sum of time spent in a car and time spent in a bus.

The families were instructed to report all occasions on which the child did not wear the personal badge, and in that case, to keep separate diaries for the child and the badge. Variables for activities noted in the diary were based on the diary of the badge to make sure that they corresponded to the personal measurement. If omissions or uncertainties were

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found in the returned diaries, we contacted the family within 1 week. Two people independently keyed in the information from the diaries, and a third person examined any differences. The time spent on each activity was calculated as the percent of the observation time, which was equal to the duration of the exposure of the child's badge.

Laboratory analysis. NO₂ was collected on the badges with triethanolamine as the substrate, which absorbs nearly 100% NO₂ and converts it to nitrite (19). The nitrite was analyzed on a segmented flow analyzer using Saltzman's reagents (20), followed by spectrophotometric detection at 540 nm. The amount of nitrite on the badges was converted to a mixing ratio by the constant of Yanagisawa and Nishimura (19). In each analytical run, two seven-points standard curves were used to determine the amount of nitrite. Furthermore, two series of four control standards were analyzed to ensure that the analytical run had proceeded properly. All control standards were prepared independently in the laboratory. At least three unexposed badges from each production series were analyzed, and the average was used as the zero-point. The detection limit for a 1-week average was 0.4 ppb NO₂.

Annual international intercalibration showed that the uncertainty of the analysis of nitrite standards was within 5% (21). On the basis of six pairs of field replicates, the coefficient of variation (standard deviation divided by mean) was estimated to be 4%; intercomparison with a TECAN chemiluminescence instrument (TECAN CLD 770 AL; TECAN AG, Hombrechtikon, Switzerland), equipped with a photolytic NO₂ converter and placed in a rural area with little pollution, showed differences within 10%. In accordance with these results, the accuracy of the method has been estimated to be within 20% (19). We also tested the badge method against two chemiluminescence NO₂ analyzers with molybdenum converters in urban areas. All analyzers were calibrated with certified permeation tubes of NO₂. When the badges were placed at the air intake of the monitoring stations, they showed significantly lower concentrations than the chemiluminescence analyzers (Table 1). The difference was expected, because several minor NO₂ compounds such as peroxycarb nitrate and HNO₃ are measured as NO₂ in the NO₂ analyzers.

Statistical methods. The relationship between the personal measurements (outcome variable) and the front-door measurements (explanatory variable) was analyzed in univariate linear regression analyses. Each residential region was analyzed separately. The analyses were based on the GLM procedure of SAS (SAS Institute, Cary, NC) (22).

The relationships between the personal measurements and the explanatory variables sex, age, traffic density at the address, NO₂ at the bedroom, NO₂ at the front door, and all variables from the diary were analyzed by multiple regression analysis (23). Multiple regression analysis was used to identify factors that influence the personal exposure of children, after correction for the other factors. The analysis was based on a mixed linear model with a region-dependent variance (the MIXED procedure of SAS) (24), as the residual variation was much larger among the children in Copenhagen than among the children in the rural areas. In the multiple analysis, the variables being outdoors, being in a city, riding in a car, riding in a bus, and riding in a car or a bus, were allowed to interact with residential region. We reduced the multiple model by successive exclusion of insignificant variables.

All tests in the multiple model were based on the likelihood ratio test statistic and those in the univariate analysis on the t-test statistic. p-Values refer to tests of no association between the explanatory variable(s) and the outcome variable.

Results

Participants and summary statistics. Of the 1,730 families invited to participate in the study, 204 were included (Table 2). Seven of the remaining 204 children were excluded because of missing values for outcome variables (exposure of the personal badge), loss of the badge (n = 3), destruction of the badge in a laundry machine (n = 1), not wearing the badge at all (n = 1), and errors in laboratory analyses (n = 2). One further front-door badge was vandalized, one indoor measurement was excluded by the laboratory because of an unrealistically low value (lower than any blank value), and one diary was excluded because of insufficient quality. The 197 children with valid exposure measurements consisted of 56 boys and 42 girls in Copenhagen and 49 boys and 50 girls in the rural districts. In Copenhagen, the majority of the homes were apartments

| Analyzer | Badge | Ratio | Difference |
|----------|-------|-------|------------|
| 12.8⁶ | 11.3 | 0.88 | 1.5 |
| 16.9⁶ | 13.0 | 0.77 | 3.9 |
| 25.3⁶ | 22.3 | 0.88 | 3.0 |
| 28.3⁶ | 25.1 | 0.89 | 3.2 |
| 31.0⁷ | 26.3 | 0.85 | 4.7 |
| 32.1⁷ | 27.5 | 0.86 | 4.6 |

*Significantly different from 1 (p = 0.0008); t-test of log(ratio) = 0.

Figures 1. Box-and-whisker plots of 195 sets of NO₂ measurements in Copenhagen and rural districts. Each set includes one measurement outside the front door, one personal measurement, and one measurement in the bedroom of the child. The box encloses the middle half of the data, and a horizontal line bisects the box at the median. The lower whisker ends at the 5th percentile and the upper whisker at the 95th percentile. Values beyond these percentiles are shown separately.
in old four- to six-story buildings, but new apartments and one-family houses were also represented. Most of the families in the rural areas lived in one-family houses with gardens.

Figure 1 shows the levels and distributions of the NO₂ measurements as box-and-whisker plots. The front-door concentrations, indoor concentrations, and personal measurements were substantially higher in Copenhagen than in the rural districts. Moreover, Figure 1 shows a decreasing trend in the NO₂ concentrations from front-door measurements through personal measurements to the bedroom measurements both in Copenhagen and in the rural areas. This trend was consistent throughout the 7 weeks during which measurements were performed (data not shown). The variability in NO₂ concentrations was greater in Copenhagen than in the rural districts, and one exceptionally high indoor concentration was found in Copenhagen.

Table 3 shows the distributions of age, traffic density, and activities as noted in the diary for each personal badge. As expected, the traffic density and the time spent in a city were much greater in Copenhagen than in the rural districts. In the rural districts, the median time spent in a car was almost twice that in Copenhagen, but the opposite situation was seen for riding in a bus. Use of gas appliances was considerably more common in Copenhagen, and exposure to passive smoking was slightly more common in Copenhagen. Riding on tractors and exposure to perceptible air pollution were rare and were therefore not considered in the further analyses. The remaining variables were similarly distributed in Copenhagen and in the rural areas. Of the 196 children with valid diary notes, 121 (62%) were not separated from their personal badges at all; the other 75 children (38%) were separated from their badges for an average of 5.1% of the observation time, and only four children were separated from their personal badges for more than 15% of the time.

Despite the substantially lowered number of homes with gas appliances and resident smokers due to the sampling strategy (Table 2), many children in this sample were exposed to three potential indoor sources of NO₂: gas appliances, passive smoking, and fire (Table 3). For example, 10% of the children in Copenhagen were exposed to passive smoking for at least 8% of the observation time (2 h/day).

### Table 3. Distribution of age, traffic density, and activities

|                     | Copenhagen children | Rural children |
|---------------------|---------------------|---------------|
|                     | Median 10th percentile 90th percentile | Median 10th percentile 90th percentile |
| Age (years)         | 8.0 4.4 11.4 | 8.8 5.0 12.2 |
| Traffic density (vehicles/day) | 10,700 1000 19,800 | 75 10 400 |
| Being outdoors*     | 10.8 5.9 21.5 | 12.4 5.1 25.2 |
| Riding in a car*    | 0.7 0.0 2.3 | 1.6 0.6 3.4 |
| Riding in a bus*    | 0.3 0.0 1.8 | 0.1 0.0 0.9 |
| Riding on a tractor* | 0.0 0.0 0.0 | 0.0 0.0 0.0 |
| Riding in a car or bus* | 1.2 0.0 3.3 | 2.0 0.6 3.9 |
| Being in a city*    | 100.0 98.6 100.0 | 1.7 0.0 6.4 |
| Passive smoking*    | 1.8 0.0 8.0 | 0.8 0.0 5.6 |
| Gas appliances used at home* | 0.0 0.0 3.1 | 0.0 0.0 0.0 |
| Near fire*          | 0.8 0.0 6.3 | 1.2 0.0 4.4 |
| Perceptible air pollution* | 0.0 0.0 0.0 | 0.0 0.0 0.0 |

*Time spent on the activity in percent of observation time.

![Figure 2](image-url)  
**Figure 2.** Plot of personal NO₂ measurements against NO₂ measurements at the front door in rural districts (n = 46) and in Copenhagen (n = 32). The median values for urban children are indicated by dotted lines.

We used the data shown in Figure 2 as an example of the potential epidemiological use of front-door concentrations of traffic exhaust fumes for classifying children into two exposure groups. Children whose front-door concentrations were below the median were classified as having low exposure and the other half of the children were classified as highly exposed. The actual exposure status was obtained from personal exposure measurements; children whose personal exposures were below the median were considered actually to have low exposure and the other half of the children actually to be highly exposed. Both the sensitivity (the proportion of correctly classified highly exposed) and the specificity (the proportion of correctly classified low exposure) of this classification method were 81% in Copenhagen and 74% in rural districts.

### Multiple analysis

The multiple regression analysis was based on 194 observations without missing values, and the initial model included 13 explanatory variables and four interaction terms. First, we...
Tables and figures refer to the study of Nitrogen dioxide exposure of Danish children. The tables compare measures such as NO$_2$ levels and traffic density across different regions, including urban and rural areas. The figures illustrate plots of observed personal NO$_2$ exposure against NO$_2$ exposure predicted by the multiple regression model in rural districts and in Copenhagen. The data show how exposure varies with time spent in urban areas vs. rural areas and the impact of traffic density and location on NO$_2$ exposure.

The discussion section delves into the implications of these findings, highlighting the importance of considering both indoor and outdoor sources of NO$_2$. The study emphasizes the need for detailed exposure assessment methods that can accurately capture the complex interaction between various factors affecting air quality in urban environments.

The text also mentions the use of advanced dispersion models to predict outdoor concentrations of NO$_2$ in streets, considering factors such as traffic density, street width, and building configuration. In Denmark, where the study was conducted, the availability of such information is crucial for effective urban planning and public health interventions.
front-door concentrations accounted for 49% of the variation in personal exposures in Copenhagen and 45% in rural areas. As an example of the use of front-door concentrations for exposure classification, we found the sensitivity and the specificity to be 81% for children in Copenhagen and 74% in rural districts. It is obvious that use of this classification method would lead to misclassification of the exposure status of children and that the misclassification (given that it is nondifferential) would lead to underestimation of the true relative risk (26,27). Nevertheless, sensitivities and specificities for classification methods of about 80% are probably not exceptions in epidemiological research.

Factors related independently to exposure to NO₂. The multiple analysis showed that the NO₂ concentrations in the bedroom and at the front door significantly affected the NO₂ exposure of Danish children, which is consistent with findings in previous studies of personal NO₂ exposure (28-33). The effect of being outdoors and in a city was also not a surprise, as substantially higher concentrations were present outdoors than indoors and in Copenhagen in comparison with rural districts.

Gas appliances have repeatedly been identified as an important source of personal NO₂ exposure (28,29,32-38). We found an effect of only borderline significance, probably due to several causes. First, most of the effect of gas appliances was included via the effect of the NO₂ concentration in the bedroom; removing the bedroom concentration from the regression model decreased the p-value for gas appliances to <0.0001. Second, the highest NO₂ concentrations have been reported from gas stoves with continuously burning pilot lights (28,39,40), which are rarely used in Denmark. Third, in contrast to most previous studies, we dealt only with children who might be expected to be less exposed than adults because 1) NO₂ concentrations in the kitchens and living rooms of houses with gas stoves decrease with decreasing height (41) and 2) children spend less of the time than their mothers do in the kitchen (36). Fourth, in the diary we asked about use of gas appliances at home, not near the child, which would tend to dilute the effect on the personal badge. Finally, the selection of the participants reduced the number of homes with gas appliances.

In the present study, we found that exposure to passive smoking increased the NO₂ exposure of children. Passive smoking has not previously been reported to increase personal NO₂ exposure, but the finding seems reliable because several studies have shown that smoking increases indoor NO₂ concentrations (39,42-44).

We found a significant effect of being near fire and particularly near burning candles. Candles are frequently used in Denmark, on the dinner table, at social gatherings like birthdays, or just to feel comfortable. In this sample, 10% of the children were exposed for more than about 1 hr/day. To our knowledge, the association between burning candles and NO₂ concentrations has not been studied previously, but an effect of burning candles seems likely due to formation of NO₂ during the combustion process.

We expected that riding in cars and buses would increase the exposure of children to NO₂, as we assumed that elevated levels on the streets would be reflected inside vehicles. Riding in cars and buses increased the exposure of rural children but decreased that of Copenhagen children. We believe that the observed effects are related to the selection of the children: the rural children lived far away from traffic and other sources of NO₂ pollution so almost any travel by car or bus would bring them to more polluted areas, whereas riding in a car or bus would often bring the Copenhagen children to less polluted areas. Moreover, the decrease of concentrations with the distance from the street is much less pronounced for NO₂ than for primary air pollutants from the traffic such as nitric oxide, carbon monoxide, and benzene (45,46). The fact that the variables riding in a car or bus and being in a city in the multiple model were at least partly competitive supports this point of view.

The results indicated that older children and girls experienced higher NO₂ exposure. If these findings are not artefacts, they must be related to some behavior of the children that was not included in the multiple model. One explanation for the effect of age and being a girl could be that older children and girls are often more competitive than younger children and boys and might therefore have followed the instructions about wearing the badge outside the clothes more accurately, thus increasing the measured concentration. This explanation is speculative, however, and we consider the results as chance findings until confirmed in future studies.

Validity of findings. Field replicates and comparisons with other measurement methods indicated acceptably low uncertainty of the badge measurement method, and the diary notes indicated that the children and families followed the instructions. Therefore, we believe that the measurements reflect the actual NO₂ concentrations. A Danish study showed small systematic seasonal changes in street concentrations of NO₂ (47) and any overestimation or underestimation of front-door levels due to random variation is probably small because we measured during 7 different weeks. Moreover, as the outdoor levels found in Copenhagen were similar to those found at a number of other locations, including Toronto, Canada (30), Watertown, Massachusetts (34), Veenendaal, Holland (36), and Middlesbrough, United Kingdom (48), we believe that the outdoor levels found in Copenhagen are representative of those in many urban locations. The outdoor levels found in rural districts are probably representative of those in areas with no major local sources of NO₂ pollution. The selection of participants, which reduced the number of children living in homes with resident smokers and gas appliances, certainly reduced the average indoor and personal exposures with respect to those in random samples in similar residential areas.

The results for a relationship between front-door concentrations of traffic exhaust fumes and personal exposures were based on the assumption that NO₂ is a marker only of traffic pollution. This assumption was strengthened by exclusion of respondents on the basis of potential indoor sources of NO₂ and secondary exclusion of participants on the basis of diary notes about the duration of children's exposure to the same indoor sources. It was not possible to avoid exposure to indoor sources entirely because very few children reported no such exposures. These exposures tend to weaken the association between front-door levels and personal exposures and, compared to the ideal situation with no indoor sources, we would expect the results of this study to be underestimates of the association. In Copenhagen, it is reasonable to consider NO₂ at the front door as a marker of traffic pollution because traffic is the dominant source of NO₂ in the streets. That is not the case in the rural districts, where the results cannot be assigned to any specific local source of outdoor NO₂ pollution.

The multiple regression analysis was based on the full sample of children. The low proportion of children exposed to indoor sources would tend to diminish the possibility of significant results for those indoor sources, but it would not discredit the significant findings. The estimates derived from a regression analysis are valid only within the range of the explanatory variables. The full sample of children did include homes with indoor sources of NO₂, and the exposure to indoor sources of the children living in those homes were probably similar to the exposure of other children with the same indoor sources. Thus, the ranges of the explanatory variables in the multiple analysis were probably similar to those in a random sample (though the averages would be higher in a random sample), and the results can probably be generalized.
without restrictions related to the sample strategy.

Our study showed a highly significant relationship between front-door concentration and personal exposure to NO₂ in a selected sample of Danish children. In the context of epidemiological studies of traffic pollution, the results indicate that the front-door concentration might be used to classify the personal exposure of urban children, although it would imply misclassification that cannot be ignored. Any major benefit of this classification method would depend on reliable dispersion models that can substitute for measurements at the front door. In most rural areas, the method would be irrelevant because of the negligible traffic density. Moreover, the study shows that passive smoking and the burning of candles increase personal exposure to NO₂, which, to our knowledge, has not been reported previously. Finally, the study confirms that indoor levels and use of gas appliances affect personal exposure to NO₂.

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