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Passive dosimeters for nitrogen dioxide in personal/indoor air sampling: A review

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Abstract

Accurate measurement of nitrogen dioxide concentrations in both outdoor and indoor environments, including personal exposures, is a fundamental step for linking atmospheric nitrogen dioxide levels to potential health and ecological effects. The measurement has been conducted generally in two ways: active (pumped) sampling and passive (diffusive) sampling. Diffusion samplers, initially developed and used for workplace air monitoring, have been found to be useful and cost-effective alternatives to conventional pumped samplers for monitoring ambient, indoor and personal exposures at the lower concentrations found in environmental settings. Since the 1970s, passive samplers have been deployed for ambient air monitoring in urban and rural sites, and to determine personal and indoor exposure to NO_2. This article reviews the development of NO_2 passive samplers, the sampling characteristics of passive samplers currently available, and their application in ambient and indoor air monitoring and personal exposure studies. The limitations and advantages of the various passive sampler geometries (i.e., tube, badge, and radial type) are also discussed. This review provides researchers and risk assessors with practical information about NO_2 passive samplers, especially useful when designing field sampling strategies for exposure and indoor/outdoor air sampling.

Keywords

nitrogen dioxide; passive sampler; diffusive sampler; personal exposure; indoor sampling

Introduction

Nitrogen dioxide (NO_2), a criteria air pollutant as defined by the US Clean Air Act[1] has been sampled in ambient and indoor air for both compliance purposes and exposure characterizations using active pumped systems as well as diffusive samplers of various designs, including badges and tubes. Nitrogen dioxide concentrations in personal air have

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[1] US Clean Air Act Amendments 1990, available at http://www.epa.gov/air/caa/ (accessed September 2007).

been typically measured using diffusive samplers because they are: (1) small in size and light weight, (2) unobtrusive and thus more readily acceptable to study participants, (3) comparatively easier to use and handle in field studies because they do not require power (e.g., battery or external electrical power), and (4) cost-effective. However, diffusive samplers have lower equivalent sampling rates than active methods and usually require relatively longer sampling times (24 h or longer). Consequently, diffusive samplers including those used for NO_2 monitoring provide integrated but not short-term concentration measurements.

Active air sampling with a pump can collect larger volumes of air and thus detect the lower concentrations found in community environments within relatively short time periods. Automated active sampling methods have been the preferred method used to monitor NO_2 continuously at ambient sites for environmental regulatory compliance activities. However, practical considerations impede the use of these continuous monitors in residential air and exposure monitoring studies. Small, low flow active samplers using battery-operated pumps have been used in a limited number of indoor and personal exposure studies.

The first passive sampling devices for NO_2, Palmes tubes, were introduced for occupational exposure monitoring (Palmes et al., 1976), but were later adapted for environmental monitoring purposes. Later, this sampler together with other tube, badge-type (Yanagisawa and Nishimura, 1982) and radial (Cocheo et al., 1996) diffusive samplers have been employed as monitors in exposure studies worldwide. There are currently several commercially available samplers, which are modifications of the original Palmes tube design. Most modifications are directed at reducing effects related to meteorological conditions (e.g., insufficient or too high a wind speed, humidity, temperature), increasing sampling uptake rates, and improving analytical sensitivity. In general, diffusive samplers have been tested and evaluated under controlled laboratory conditions and, less frequently, in the field, although not in a fully comprehensive manner.

The focus of this review is to present a critical summary of the evaluation of passive sampling methods, because these methods are currently the most reliable and feasible way for measuring personal NO_2 exposure. This review provides useful and practical information for researchers when designing population-based studies of nitrogen dioxide concentrations in outdoor, indoor and personal air. Limitations and measurement artifacts are described as they are especially germane to the use of passive samplers in exposure studies. When independent evaluations of a passive sampler performance have not been done but the monitor appears to present potential advantages over existing samplers, data reported by the monitor manufacturers are included along with caveats concerning the interpretation of data collected with the passive sampler.

NO_2 passive sampler modifications

Geometry

Passive sampler designs are based on the well known diffusion principle described by Fick's law (Krupa and Legge, 2000). Strictly, Fick's law applies only under ideal, steady state conditions assuming that the sorbent is a perfect sink. Since there can be deviations from ideal conditions in actual sampling, the theoretical sampling rate for a given analyte and the actual rate can differ, because the latter depends on the sampler's geometry and the environmental conditions. Modifying the geometry of the diffusive sampler by reducing the diffusion path length and/or increasing the diffusion cross-sectional area of the sampler are straightforward approaches for optimizing sampling rates. However, the impact of deviations from ideality on actual sampling rates poses a limit on the extent that sampler geometry modifications are possible without incurring unacceptable deviations from Fick's law.

Analytical Sensitivity

NO_2 passive samplers are typically chemisorptive, that is NO_2 reacts with the chemically-treated sorbent which is extracted post-sampling and the reaction derivatives are chemically analyzed in the extract. The mass of NO_2 collected is estimated from the concentration of the derivative(s) in the extract based on the stoichiometry of the reaction. Therefore, one approach to improving detection limits associated with passive sampling measurements is to optimize the chemisorptive reaction and the extraction/analysis procedures to increase analytical sensitivity. The most commonly used NO_2 passive samplers rely on the classical reaction with triethanolamine (TEA). However, there are instabilities in the formations of the NO_2-TEA adduct, that have led to alternative approaches to the formulation of the sorbent and extraction/analysis procedures. Further, while chemisorption is less prone to the back diffusion phenomenon than adsorptive-only passive samplers, artifact analyte losses can still occur due to interferences from other pollutants that also react with the sorbent or the derivatives. Known interferences include nitrous acid, peroxy acetyl nitrate and nitric acid (Gair et al., 1991).

Designs

Since the development of the initial NO_2 passive sampler, the Palmes tube, passive samplers have been modified by changing the sampler's physical dimensions and chemical components. The samplers are generally classified into three major designs: tube, badge, and radial types. Tube-type samplers are characterized by a long, axial diffusion length, and a relatively small cross-sectional diffusion area, which results in relatively low sampling rates (Namiesnik et al., 2005). Badge-type samplers have a shorter diffusion path length and a greater cross-sectional area which results in sampling rates that are typically higher than diffusion tubes (Namiesnik et al., 2005); but the sampling rate may be more variable and deviate from the theoretical rate because badge samplers are affected by turbulence more. With one exception, tube- and badge-type passive samplers are based on an axial design. This design operates by diffusion in the axial direction and cannot attain higher sampling rates, especially in the case of narrow-tube geometries (Cocheo et al., 1996). This limitation is addressed by the radial-type sampler design (Namiesnik et al., 2005). The radial design provides for a higher diffusive surface without an increase in amount of adsorbing media compared to axial design. The physical characteristics of passive samplers (tube-type, badge-type, and radial type) are summarized in Table 1. Passive sampler performance characteristics are listed and compared with typical active samplers in Table 2.

Tube designs

Palmes Tube

The Palmes tube was developed initially as a personal monitoring device for SO_2 and water vapor, and later adopted for workplace monitoring of NO_2, especially for underground mine workers (Palmes et al., 1976). Subsequently, the application was extended to indoor air monitoring (Atkins et al., 1978). It is constructed from a length of acrylic tube, three stainless steel screens coated with TEA that are stacked at the bottom of the tube, and two polyethylene caps for fixing the screens and protecting the open inlet of the tube (Palmes et al., 1976). The TEA-NO_2complex formed is extracted post sampling and analyzed by UV/VIS spectrophotometry at 545 nm. To improve the analytical sensitivity and precision of analysis, ion chromatography is now preferred to the traditional spectrophoto-metric method. The two detection techniques provide comparable results (Gair et al., 1991; Gerboles et al., 2003). A modification of the traditional Palmes tube has been made by fitting a membrane at the open end of the tube to minimize the effect of turbulence due to varying wind speed (Gerboles et al., 2005). This sampler has been commercialized by Gradko Environmental and is frequently referred to as the Gradko sampler in technical reports.

For environmental applications, the Palmes tube requires week-long sampling periods. It has been extensively used for residential indoor and indoor/outdoor measurements, for exploring the relationship between indoor and outdoor levels (Cyrys et al., 2000; Janssen et al., 2001; Raw et al., 2004; Simoni et al., 2004), for measurement of NO_2 in ambient air (Glasius et al., 1999; Stevenson et al., 2001; Lewne et al., 2004; Gauderman et al., 2005; Gonzales et al., 2005; Da Silva et al., 2006), and in more limited fashion, for personal exposure studies (Mukala et al., 1996; Chao and Law, 2000; Kousa et al., 2001; Lai et al., 2004). Some studies have evaluated the reliability of this passive sampler's performance during field studies by comparison to chemiluminescence analyzers (Gair et al., 1991; Gair and Penkett, 1995; Kirby et al., 2001; Plaisance et al., 2004). The majority of these studies indicate that while these samplers have very good precision (generally within 5%) they tend to overestimate NO_2, by 10 to 30% as compared to chemiluminescence analyzer measurements, suggesting interferences from other reacting species.

The Palmes tube was the first passive sampler developed for measuring NO_2 in the air, and has been the most extensively used to date, especially in Europe, as an alternative ambient air monitor. The most appropriate application for this sampler is for ambient air monitoring with a sampling duration of 1 month. As with other passive dosimeters, it does not capture short-term, fluctuating NO_2 concentrations, which are often important for exposure and health assessments.

Passam Sampler

This sampler is derived from the design of the Palmes tube (Gerboles et al., 2006). The sampler is designed for two different sampling durations, week-long (longer-term) and 8–48h (shorter-term). The longer-term Passam sampler is slightly cone-shaped with a diffusion path length of 73.5 mm and inner diameter of 9.82 mm. The shorter-term Passam sampler has a shorter diffusion path length of 20 mm and a larger diffusion diameter of 23 mm. The sampler has been found to perform well at a range of meteorological conditions: R.H. between 20–80%, temperature between 5–45°C, and wind speed between 0.1–4.0 m/s (Hangartner, 2001). Sampling rates were evaluated in laboratory experiments and were found to be comparable to those estimated from field studies performed in Switzerland (Passam instruction[2]).

The shorter-term Passam sampler is applicable for daily measurements to monitor air quality standards or personal air concentrations in exposures studies. The longer-term Passam sampler was used to measure two-week averaged personal NO_2 levels for children in Oslo (Magnus et al., 1998), and in an ecological study for determination of NO_2 in ambient air (Buffoni, 2002).

Analyst™ Passive Sampler

The Analyst™ passive sampler is also a modification of the Palmes-tube design. It was originally developed for monitoring aromatic volatile hydrocarbons in ambient air (Bertoni et al., 2001) and later modified to determine NO_2 and NO_X (De Santis et al., 2002). The body of the sampler is a cylindrical glass vial with a threaded cap at one end. NO_2 is collected on a disk of carbon paper filter impregnated with a 1% (w/v) sodium carbonate + 1% (w/v) glycerin solution, and placed at the bottom of the vial and held in position by a stainless steel ring (De Santis et al., 2002). For NO_X collection, a carbon paper filter impregnated with NO_X oxidizing CrO_3 or $K_2Cr_2O_7$ is used. After sampling, the filter is extracted and analyzed by ion chromatography.

This sampler is suitable for long-term monitoring (typically one month) of oxides of nitrogen, sulfurdioxide, and volatile organic compounds in ambient air. The Analyst™ passive sampler is a reliable tool for long-term determination of concentrations in indoor and outdoor environments (Bertoni et al., 2001). The sampler can be used as a screening tool for ambient monitoring to identify pollution “hot spots” where concentrations are likely to be consistently high (De Santis et al., 2004). As with diffusive samplers in general, these monitors can provide information on spatial distribution of concentrations at lower cost than active samplers.

Badge designs

Yanagisawa Filter Badge

Badge-type, NO_2 passive samplers were developed to provide higher diffusion rates and reduced sampling duration compared to Palmes tubes (Yanagisawa and Nishimura, 1982) so they are more suitable for personal exposure measurement at environmental concentrations. The Yanagisawa filter badge is comprised of three parts: an absorbent sheet coated with TEA, a diffusion fiber filter with a pore size of 5 μm, and a badge case (Yanagisawa and Nishimura, 1982). This sampler was successfully validated in the laboratory over a range of wind velocities (0.15–4.0 m/s) and relative humidities (40–90%) with a 7-day exposure. The filter badge has been optimized and evaluated for indoor environments and personal

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[2] Passam instruction: Sampling rate of NO_2 sampler, available at http://www.passam.ch/information.htm (accessed July 2007).

monitoring (Lee et al., 1993a, b) and extensively used for personal exposure studies (Yanagisawa and Matsuki, 1986; Berglund et al., 1994; Ramirez-Aguilar et al., 2002; Lee et al., 2004), indoor measurements (Smedje et al., 1997; Shima and Adachi, 2000; Kodama et al., 2002; Algar et al., 2004; Bae et al., 2004), as well as for ambient air monitoring (Norris and Larson, 1999; Tashiro and Taniyama, 2002; Levy et al., 2006). Owing to the greater uptake rate resulting from the larger cross-sectional area of the badges and shorter diffusion length than tube-type samplers, sampling times can be as short as one day at typical environmental air concentrations.

Ogawa Sampler

The Ogawa sampler is a double face badge that can simultaneously monitor NO, NO_X and NO_2 (Ogawa protocol[3]).The sampler is cylindrical and comprised of two end chambers (Ogawa protocol). Each chamber contains a collection filter, pre-coated with specific absorbent(s) and placed in between two screens. The coated filter and screens are secured by a patented diffuser end cap which is composed of a Teflon disk with 2 mm diameter holes (Tang et al., 2001). For simultaneous collection of NO_X and NO_2, the NO_2 collection filter is coated with TEA and the NO_X collection filter is coated with TEA and 2-phenyl-4,4,5,5-tetramethy-limidazoline-1-oxyl-3-oxide, an oxidation reagent, added to convert NO to NO_2, are set in each of the two chambers.

Ogawa samplers have been extensively used in human exposure studies to measure personal air concentrations and residential indoor/outdoor levels across a range of populations and in multiple locations, including adults of Richmond, Virginia (Zipprich et al., 2002), children of Santiago, Chile (Rojas-Bracho et al., 2002), office workers of Paris, France (Mosqueron et al., 2002), and cardiac disease-compromised individuals in Toronto, Canada (Kim et al., 2006). The samplers also have been used in ambient air monitoring networks to assess traffic-related pollutant exposures (Singer et al., 2004) as well as to evaluate spatial variability of nitrogen dioxide ambient concentrations in Montreal, Canada (Gilbert et al., 2005).

The Ogawa sampler is useful for conducting ambient air pollution monitoring. The sampler is cost effective since: (1) it is feasible to measure two target air pollutants simultaneously by using two different kinds of collection filters and (2) it is reusable countless times since only the pre-coated collection filters are replaced. Good agreement was obtained between these passive devices and chemiluminescence analyzers in two field comparison studies with r^2 of 0.91 for 16 paired measurements (Mukerjee et al., 2004) and r^2 of 0.95 for 107 paired measurements (Sather et al., 2007).

IVL Sampler

Tube-type (Sjodin et al., 1996) and badge-type (Ferm and Svanberg, 1998) passive samplers have been developed by IVL (Swedish Environmental Research Institute). A field evaluation for nitrogen dioxide monitoring in urban air was conducted by Royset (1998)

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[3] Ogawa protocol: NO, NO_2, NO_X and SO_2 sampling protocol using the Ogawa sampler, Edition 6.06, June, 2006, available at http://www.ogawausa.com/protocol.html (accessed July 2007).

using a continuous NO/NO_X monitor, an active (pumped) glass tube encasing a collection filter pretreated with sodium iodide, and an IVL badge-type sampler. Results indicated that the agreement between the IVL badges and the conventional active sampling methods was approximately 20%, with r^2 of 0.78 and 0.79, respectively, with active tube and continuous monitor measurements.

The badge-type IVL sampler is suitable for air monitoring over sampling durations from 1 to 3 months (IVL brochure[4]). Manufacturer-reported detection limits for this sampler with sampling times of ~1 month are 0.1 and 0.5 $\mu g/m^3$ for NO_2 and NO, respectively. Owing to its long sampling time, this sampler has been extensively used for NO_2 background monitoring in ambient air at rural or remote sites (Ferm and Rodhe, 1997; Pleijel et al., 2004).

Willems Badge

The Willems badge was developed at the University of Wageningen, Netherlands, and originally intended for airborne ammonia measurements and later modified for measuring NO_2 (Hagenbjork-Gustafsson et al., 1996). It consists of a cylinder of polystyrene with a Whatman GF-A glass fiber filter impregnated with triethanolamine at its base and held in place by a 6 mm distance ring. A Teflon filter is placed on the 6 mm polystyrene ring, which is secured with a polystyrene ring of 3 mm. The badge is closed by a polyethylene cap.

This passive sampler has been used for monitoring ambient NO_2 concentrations with short-term sampling (e.g., 24 h) when the wind speed is ≥2 m/s (Hagenbjork-Gustafsson et al., 1999). It is also appropriate for personal sampling in working environments at a minimum wind velocity of 0.3 m/s (Hagenbjork-Gustafsson et al., 2002). This badge sampler was used to measure NO_2 concentrations inside and outside of homes of asthmatic children residing in Europe (Hagenbjork-Gustafsson et al., 1996) and individuals working in office buildings in Umea, Sweden (Glas et al., 2004).

Krochmal Badge

The Krochmal badge-type sampler was first introduced in 1983 as the Amaya-Sugiura passive sampling method for NO_2 concentration measurements in ambient air (Krochmal and Gorski, 1991a), and then modified and evaluated under a range of meteorological conditions (temperature and humidity) as well as tested for storage stability and different TEA carriers (Krochmal and Gorski, 1991b). The sampler was further modified to measure ambient NO_2 and SO_2 simultaneously by using a black stained polythene sampler body to protect the absorbing pad from sunlight (Krochmal and Kalina, 1997a).

Nitrogen dioxide and sulfur dioxide were measured in urban and rural areas of Poland for a 1-month sampling period (Korchmal and Kalina, 1997b). The measurement sites were divided into five different categories, that is, centers of cities, residential areas, industrial areas, traffic locations, and rural areas. The measurement results were consistent and reliable at the various geographical areas tested. Further work is needed to demonstrate the

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[4] IVL brochure: Diffusive samplers for air monitoring, available at http://www3.ivl.se/affar/miljo_kartl/proj/passive_sampl/PassivaProvtagare.pdf (accessed July 2007).

applicability of this badge for monitoring low concentrations of NO_2 and SO_2 over 24-h sampling periods (Krochmal and Kalina, 1997a).

Maxxam PASS

The Maxxam PASS badge-type sampler was first introduced for measuring SO_2 in ambient air (Tang et al., 1997) and modified to measure NO_2 for month-long air monitoring (Tang et al., 1999). This sampler employs a new, proprietary chemically pre-treated solid collection medium, CHEMIX™, instead of the conventional triethanolamine-coated sorbent matrix. Given that the exact chemical composition of the chemisorptive adsorbent cannot be openly accessed, and that field evaluation data are limited, the information provided on the performance of this passive sampler should be carefully considered. The manufacturers' reported advantages of this new collection medium are (1) lower sampling rate reduction (10 vs. 80%) with a temperature drop from 20 to −27°C, (2) higher sampling rate (0.97 vs. 0.19 μg/ml as collected nitrate concentration), and (3) greater stability in a field evaluation test (Tang et al., 1999). The ambient NO_2 dynamic concentration range was reported as 0.1–50 ppb over a 1-month exposure period by Tang et al. (1999). The use of this sampler for personal monitoring has not been reported.

Radial designs

Radiello® Sampler

The Radiello sampler uses radial diffusion over a microporous cylinder into an absorbing inner cylinder, instead of axial diffusion. The radial design results in up to 100-fold increase in sampling rate (Hertel et al., 2001) compared to axial diffusion. Sample collection from 1 to 15 days is feasible but relative humidity higher than 70% can cause interferences when used for extended periods of more than 7 days (Radiellomanual[5]).

This radial diffusive sampler was used to measure personal exposures and residential indoor/outdoor air concentrations over 48 h in Copenhagen, Denmark (Sorensen et al., 2005), and for 1 week-long monitoring in a Mediterranean coastal area of Spain (Delgado-Saborit and Esteve-Cano, 2006).

EMD (Ecole des Mines de Douai) Sampler

A new high-sampling rate diffusive sampler has been recently developed and evaluated in the laboratory and in the field for measurement of NO_2 levels in ambient air (Piechocki-Minguy et al., 2003). It is composed of a porous cartridge impregnated with triethanolamine and fitted in a cylindrical protective box equipped with caps at its extremities (Piechocki-Minguy et al., 2006). The cylindrical configuration provides a high surface area with a corresponding higher sampling rate but limited extraction volume (Piechocki-Minguy et al., 2003). The sampling rate was reported to be an average of 0.89 cm^3/s for indoor sampling and 1.00 cm^3/s for outdoor sampling. The sampling rate was not significantly influenced by wind at speeds higher than 0.3 m/s (Piechocki-Minguy et al., 2003).

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[5] Radiello manual: English Edition v.01-2006, available at http://www.radiello.com/english/Radiello'smanual 01-06.pdf (accessed July 2007).

A field comparison between EMD samplers and chemiluminescent NO_2 monitors showed strong correlation between the samplers ($r = 0.90$) at three locations (two suburban and one curbside station) with 4-h sampling periods during both summer and winter (Piechocki-Minguy et al., 2003). This device was used in sampling campaigns to assess personal exposures in a series of microenvironments (home, other indoors, transport and outdoor) for two 24-h long periods (weekdays and weekends) (Piechocki-Minguy et al., 2006). Owing to its higher sampling rate, this passive sampler may be useful for measuring short-term personal exposure to NO_2, perhaps even for 1-hour periods. However, further validation and optimization for hourly measurements of NO_2 concentrations are necessary before applying them for short-term personal exposure studies.

Comparison and evaluation of NO_2 passive samplers

Sampler Types

The applications, limitations, and extent of independent evaluation for each passive sampler described in this review are summarized in Table 3. Tube-type passive samplers requiring sampling durations of at least 1 week up to 1 month have been evaluated for ambient air monitoring. Because of the relatively long-duration sampling requirements, tube-type samplers have limited utility for personal air monitoring and cannot be used to characterize short-term variability in exposure, which is a critical need in health effects studies. Badge-type passive samplers have higher sampling rates and can measure 24-h average NO_2 concentrations. However, the capability of measuring personal air with very short time (e.g., hourly sampling) is still questionable. To conduct short-term personal NO_2 exposure measurements in micro-environments (e.g., in vehicle or environments such as restaurants that are visited during relatively short time periods), a passive sampler with higher sampling rate than tube or badge-types is needed. The radial type passive samplers that have been developed to enable the sampling over a range from a few hours to 1 week is promising but has not been independently confirmed.

NO_2 Absorbents and Interfering Compounds

In addition to the geometry changes directed at increasing the sampling rate, efforts to improve the analytical sensitivity have also been made to reduce the required minimum sampling duration. These modifications have included altering the analysis methods for TEA-adducts since TEA is the most common adsorbent of NO_2, and introduction of other adsorbents, such as a mixture of 1% (W/V) of sodium-carbonate and glycerin used for Analyst™ passive sampler, a combination NaI+NaOH for IVL sampler, and CHE-MIX™ for Maxxam PASS. These reagents attempt to improve the stability of NO_2 absorbed by the impregnated filter (IVL and Analyst™ passive sampler) which enhances the capability of the sampler for monitoring at environmental conditions (Maxxam PASS). However, as mentioned above, these adsorbents have not fully validated in field studies and more data are required to demonstrate that they constitute an improvement over the traditional TEA adsorbent.

Evidence indicates that there is a measurement bias in NO_2 passive sampler results compared to active monitors probably due to interfering compounds, usually other gas-phase nitrogen oxide species. Tube-type passive samplers tend to overestimate ambient air NO_2 concentrations compared to chemiluminescence analyzers. This bias appears more pronounced for the axial diffusion tube types, probably due to the longer path length and, therefore, transit time between the sample inlet and the chemisorptive surface that could enhance interaction with the interferent compounds. The sampling artifact could be explained, in part, by potential formation of NO_2 generated via chemical reactions between ozone and nitric oxide (NO) within the diffusion tube body, thus leading to an overestimate of the concentration varying from 10% (Bush et al., 2001) to as much as 30% (Campbell et al., 1994; Heal et al., 1999; Cox, 2003). Owing to spatial and temporal variability of NO and NO_2 concentrations, especially at roadsides where nitric oxide concentrations are relatively high, the presence of sufficient ozone can result in inter-conversion between these species.

Environmental Factors

Environmental factors, such as temperature, relative humidity, and wind speed can affect the performance of passive samplers (Table 1). Consideration of these environmental factors when selecting a sampler for field studies is important. The performance information reported by manufacturers may not provide these and other caveats for their use, so careful assessment of the data source provided is necessary and evaluation of performance in pilot studies is advisable before implementing full scale field sampling whenever independent evaluations in the peer reviewed literature are not available. As a general rule, tube-type samplers are typically less affected by extremes in air velocities than badge designs which are more likely to experience varying sampling rates due to shortening or lengthening of the sampler boundary layer (Brown, 2000). Radial diffusive samplers require a minimal face velocity of 0.25 m/s. High humidity can alter the sorption behavior of the adsorbent-coated surface inside a tube-type sampler, particularly if condensation occurs (Brown, 2000). If humidity is an important interferent and given the same adsorbent, a higher sampling rate device may not be optimal because it will be more prone to the humidity effect. Sampling rates determined under ideal conditions may not be valid across the varying environmental conditions that occur throughout a typical sampling period and need to be further evaluated across a wide range of concentrations, wind velocity, temperature, and relative humidity (Varshney and Singh, 2003).

Summary and recommendations

Some of the NO_2 passive samplers described have been extensively used for personal exposure measurement and indoor air sampling over one to several day time periods as well as for ambient air monitoring for up to several weeks. Recently developed passive samplers have potential for lower detection limits and utility for shorter sampling times, but need to be fully evaluated. In addition to their direct applicability in human exposure and health studies, there is also an increased need for air monitoring in remote and unpopulated areas for the purpose of evaluating ecosystem impact and to obtain data for transport model evaluation. All these passive samplers are light, easy to carry and handle by field technicians or study participants, and can be worn by young children and even toddlers. However, passive samplers have not yet provided exposure information about short-term, time-varying NO_2 concentrations and can have measurement biases due to interferences from co-existing air pollutants or environmental factors, which need to be addressed prior to field implementation in exposure assessment studies.

Geometric characteristics are major factors in determining passive sampler applicability and performance. Generally, modifications to the Palmes tube design by increasing the diffusion cross-sectional area and decreasing the diffusion path length have resulted in increases in the sampling rate. However, due to the limitations imposed by Fick's law and the practical considerations in personal monitor design, further improvements may require other approaches. A radial design for passive sampler has been recently developed as an alternative but additional laboratory and field evaluations are required, including comparison to reference active air sampling system to confirm its validity for measuring airborne NO_2 concentration under different environmental conditions.

The selection of the appropriate passive sampler is very important in designing a sampling plan for NO_2 personal and indoor air measurements. The tables summarizing the nature and characteristics of currently available NO_2 passive samplers provided in this review are useful as a first step for that purpose. Finally, as is the case with passive samplers available for monitoring airborne contaminants other than NO_2, there is a need for undertaking well designed field studies where the passive samplers are collocated with traditional sampling methods in realistic conditions, covering a range of geographic settings and meteorology conditions. Given the extensive network of fixed sampling sites with continuous monitoring for NO_2/NO_x for the purpose of compliance with the NAAQS, a thorough performance evaluation of passive devices is possible.

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Table 1

Passive samplers used in NO_2 measurements.

Passive sampler	Dimensions (diffusion length × diffusion area)	Absorbent	Analytical method	Sampling rate	
				Manufacturer	Experiment
Tube type					
Palmes tube (Gradko sampler)	7.1 cm × 0.95 cm^2	Triethanolamine	Spectrophotometry/ion chromatography	1.2 cm^3/min	1.212 cm^3/min
Passam sampler					
Short	2.0 cm × 4.15cm^2	Triethanolamine	Spectrophotometry	15.5 cm^3/min	N.R.
Long	7.4 cm × 0.75cm			0.85 cm^3/min	0.87 cm^3/min
Analyst™ passive sampler	2.54 cm × 3.27 cm^2	Na_2CO_3+$C_3H_5(OH)_3$	Ion chromatography	12.3 cm^3/min[a]	11.7 cm^3/min
Badge type					
Yanagisaw a filter badge	1.0 cm × 20 cm^2	Triethanolamine	Spectrophotometry	N.A.	5.4–6 cm^2/min[b]
Ogawa sampler	0.6 cm × 0.79cm^2	Triethanolamine	Spectrophotometry	12.1 cm^3/min[a]	9.5 cm^3/min[c]
IVL sampler	1.0 cm × 3.14cm	KI+ $NaAsO_2$ or NaI+NaOH	Spectrophotometry	29 cm^3/min[a]	N.A.[d]
Willems badge	0.6cm × 6.16cm	Triethanolamine-acetone	Spectrophotometry/ion chromatography	N.A.	46cm^3/min
Krochmal badge	1.0 cm × 4.91 cm	Triethanolamine	Spectrophotometry/ion chromatography	45.3 cm^3/min[a]	N.R.
Maxxam PASS	8.0 cm × 17.35cm	CHEMIX™	Spectrophotometry	200 cm^3/min[a]	47–68 cm^3/min
Radial type					
Radiello® sampler	1.8 cm × 2.0 cm^2	Triethanolamine	Spectrophotometry	75 cm^3/min	N.A.
EMD sampler	0.4 cm × 11 cm	Triethanolamine	Ion chromatography	N.A.	53–60 cm^3/min

Passive sampler	Known or potential	Environmental factors influences			References
		Temperature	Relative humidity	Wind speed	
Tube type					
Palmes tube (Gradko sampler)	Ozone, Nitrous acid, Peroxy acetyl nitrate	<10% between 2 and 30°C	<10% between 20 and 80%	<10% between 1.0 and 4.5 m/s	Gradko data sheet Plaisance et al., 2002, 2004
Passam sampler					
Short	Oxidants	No influence from 10 to 30°C	No influence from 20 to 80%	<10% up to 4.0 m/s	Passam instructions
Long	Peroxy acetyl nitrate	No influence from 5 to 40°C			

Passive sampler	Dimensions (diffusion length × diffusion area)	Absorbent	Analytical method	Sampling rate	
				Manufacturer	Experiment
Analyst™ passive sampler	Nitric oxide	N.A.	Unaffected from 20 to 80%	±30% from 0.2 to 2.0 m/s	De Santis et al., 2002
Badge type					
Yanagisawa filter badge	N.R.	Deviation occurs <0 or >30°C for 24-h exposure	±30% between 40 and 80%	±30% between 0 and 4.0 m/s	Yanagisawa and Nishimura, 1982 Advantec instruction
Ogawa sampler	N.R.	Operable from —10 to 40°C	Operable from 50 to 80%	N.A.	Ogawa protocol
IVL sampler	N.R.	<30% from 5 to 25°C[e]	<40% from 30 to 80%[e]	<40% from 1.0 to 4.2 m/s[e]	IVL brochure Ferm and Svanberg, 1998
Willems badge	N.R.	N.A.	Not significant from 20 to 80%	<15% from 0.3 to 2.0 m/s[f]	Hagenbjork-Gustafsson et al., 1999, 2002
Krochmal badge	N.R.	63% increase from 0 to 30°C	25% increase from 10 to 100%	N.A.	Krochmal and Gorski, 1991a, b, 1997a
Maxxam PASS	N.R.	4% increase from −27 to 20°C	<4% from 4 to 50%	≈40% increase from 0 to 1.5 m/s[g]	Tang et al., 1999 Tang et al., 2001
Radial type					
Radiello® sampler	Excess water may cause a loss of NO_2 adsorbed onto media	Not varying from − 10 to 40°C	Invariant in the range of 15–90%	Invariant between 0.1 and 10m/s	Radiello brochure
EMD sampler	N.R.	Varying ±15% from 5 to 30°C	Varying ±15% between 50 and 80%	Constant from 0.3 to 1.4 m/s	Piechocki-Minguy et al., 2003, 2006

[a] Sampling rates are theoretically obtained through the sampler's geometry and Fick's diffusion law (Tang et al., 2001).

[b] Sampling rate for Yanagisawa badge was reported as diffusion coefficient (5.4–6.0 cm^2/min), which was similar to overall mass transfer coefficient of NO_2 (6.0 cm/min) in personal monitoring and indoor environments (Lee et al., 1993a, b).

[c] Sampling rate for the Ogawa sampler was calculated from a conversion factor (56 p.p.b.-min/ng) for NO_2 at 20°C and 70%.

[d] IVL sampler do not use a fixed sampling rate to calculate a concentration (personal communication). NR and NA represent not reported and not available, respectively, in the references.

[e] The values were obtained and calculated from laboratory comparison tests for IVL sampler by Gerboles et al. (2006).

[f] Validation tests were conducted with various positions, but the values were taken from parallel to wind blowing direction in laboratory study.

[g] The percentage was calculated from an empirical equation provided in Tang et al. (1999).

Table 2

The performance of sampler/sampling method for NO_2 measurements in the air.

NO_2 sampler/monitor	Duration of sampling	Working concentration	Detection limit	QA/QC	References
Active sampling					
Impinger method	2–24h	10–400 p.p.b.	N.R.		Goyal, 2003
Chemiluminescence	Continuous	0.5–1000 p.p.b.	0.05 p.p.b.	RSD <5%	
Personal monitor	Real-time	0–50 p.p.m.	0.1 p.p.m.	Accuracy ±5%	OdaLog 6000 brochure
Passive sampling					
Palmes tube (Gradko sampler)	2–4 weeks	1.0–10,000 p.p.b.[a]	0.4 p.p.b. for 2 weeks	Uncertainty <30% above 10.6 p.p.b.; Precision <5%	Gradko data sheet Plaisance et al., 2002, 2004
Passam sampler					
Short	8–48h	2.7–128 p.p.b.	1.1 p.p.b. for 8 h 2.7 p.p.b. for 48 h	Uncertainty 28.8% at 43 p.p.b.	Passam instructions
Long	1–4 weeks	0.5–106 p.p.b.	0.2 p.p.b. for weekly 0.4 p.p.b. for fortnight	Uncertainty 23.4% at 11–21 p.p.b.	
Analyst™ passive sampler	1–3 months	13–657 p.p.b.	N.R.[b]	Accuracy ±20%; Precision ±3%	De Santis et al., 2002
Yanagisawa filter badge	1–7 days	0–19 p.p.m.*hr[c]	66 p.p.b.*hr	Accuracy ±20%; CV 4.8%	Yanagisawa and Nishimura, 1982 Advantec instruction
Ogawa sampler	<24h <168h	0–25 p.p.m. 0–3.6 p.p.m.	2.3 p.p.b. for 24 h 0.3 p.p.b. for 168 h	Differences <10%[d]	Ogawa protocol
IVL sampler	1 month +	0.05–213 p.p.b.	0.05 p.p.b. for 1 month	Agreement within ±10%; RSD 3.9%	IVL brochure Ferm and Svanberg, 1998
Willems badge	2–8h and 1–7 days	1.1–80 p.p.b.	1.2 p.p.b. for 8 h 0.3 p.p.b. for 7 days	Uncertainty 24%; Accuracy <10%	Hagenbjork-Gustafsson et al., 1999, 2002
Krochmal badge	1–30 days	0–133 p.p.b.	8.0 p.p.b. for 1 day 0.3 p.p.b. for 30 days	Accuracy 10%; RSD 6% above 11p.p.b.	Krochmal and Gorski, 1991a, b, 1997a
Maxxam PASS	1–4 weeks	0.1–50 p.p.b.	0.1 p.p.b. for 1 month	Accuracy within 15%; Precision within 5%	Tang et al., 1999 Tang et al., 2001
Radiello® sampler	1–24h and 1–7 days	1.0–500 p.p.b.	1.0 p.p.b. for 7 days	Uncertainty 11.9%	Radiello brochure
EMD sampler	1–24h	2.7–136 p.p.b.	5.8 p.p.b. for 1 h	Uncertainty 34–38% for lab test and 28–37% for field test	Piechocki-Minguy et al., 2003, 2006

[a] The level of working concentration is obtained through the measurement by dilution of sample in the laboratory experiment.

[b] The manufacturer provides a detection limit for Analyst™ sampler as 100 $\mu g/m^3$ depending upon the analytical method and target compound.

[c] Working concentrations are obtained from an agreement between the theoretical (based on reference methods) and the tested passive samplers.

[d] The value was obtained from a linear regression between Ogawa passive samplers and continuous real-time monitors. (passive sampler = 0.909 × continuous monitor+1.5029; R^2 =0.9936).

Table 3

The comparison/evaluation of passive samplers described in the paper.

Passive sampler	General use	Benefit	Weakness
Tube type			
Palmes tube (Gradkosampler)	Long-term outdoor monitoring	Reliable passive sampler by long-term use	Not suitable for short-term monitoring overestimation occurred
Passam sampler Short Long	Long/short-term ambient air monitoring	Applicable to two different sampling durations	Field applications are limited
Analyst™ passive sampler	Long-term air monitoring network	Ease of extraction NO/NO_2/NO_X simultaneous measurement	High detection limit
Badge type			
Yanagisawa filter badge	Ambient air monitoring Personal/ indoor air sampling	Long-established badge system	Not reliable for longer exposure
Ogawa sampler	Ambient air monitoring network Indoor air sampling	NO/NO_2/NO_X simultaneous determination	Limited application for personal exposure measurement
IVL sampler	Long-term ambient monitoring	Specifically for longer exposure (3 months)	Not useful for shorter exposure
Willems badge	Personal/indoor air sampling	Relatively high sampling rate	Not commercialized
Krochmal badge			
Maxxam PASS	Month-long ambient monitoring	Robust to environmental conditions	Verifications are needed in the field Not for personal exposure study
Radial type			
Radiello® sampler	Ambient air monitoring Personal/ indoor air sampling	High sampling rate	Not suitable for long-term ambient monitoring
EMD sampler	Micro-environmental exposure Measurement within hours	Operable by 1-hour sampling	Still developing Need to be verified and optimized