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INVITED ARTICLE

Optimized design of substrate-integrated hollow waveguides for mid-infrared gas analyzers

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Abstract

Design and analytical performance studies are presented for optimizing a new generation of hollow waveguides suitable for quantitative gas sensing—the so-called substrate-integrated hollow waveguide (iHWG). Taking advantage of a particularly compact Fourier transform infrared spectrometer optimized iHWG geometries are investigated toward the development of a multi-constituent breath analysis tool compatible for usage, e.g., in exhaled mouse breath analysis. Three different iHWG geometries were compared, i.e., straight, meandering one-turn and meandering two-turn waveguide channels aiming at maximizing the related analytical figures-of-merit including the achievable limits of detection for selected exemplary analytes. In addition, efficient coupling of infrared (IR) radiation into straight iHWGs was investigated using integrated optical funnel structures. Calibration functions of butane in nitrogen serving as IR-transparent matrix gas were established and compared for the various iHWG geometries. Given the tidal volume of exhaled breath (EB) samples ranging from a few hundreds of milliliters (human, swine) to a few hundreds of microliters (mouse), it is essential for any given analysis to select an appropriate waveguide geometry and volume yet maintaining (i) a compact footprint ensuring hand-held instrumentation, (ii) modular exchange of the iHWG according to the analysis requirement yet with constant device format, and (iii) enabling inline/online measurement capabilities toward continuous EB diagnostics.

Keywords: substrate-integrated hollow waveguide, mid-infrared, mid-infrared sensors, optical sensors, gas sensing, hydrocarbons

(Some figures may appear in colour only in the online journal)

1. Introduction

The range of applications that hollow waveguides (HWGs) are typically deployed in extending from medical usage and industrial process analysis to environmental monitoring confirm their versatility, as evidenced in a series of recent publications [1–4]. Initially, HWGs were predominantly designed for surgical laser power delivery systems merely using hollow-core fibers as light-pipes [5–8]. Most commonly, conventional HWGs consist of e.g., a silica structural tube with an infrared (IR)-reflecting metal layer (e.g., Ag) lining the inside wall of the hollow core protected by an IR-transparent dielectric overcoat (e.g., AgI).

Next to serving as a photon conduit with IR radiation guided by reflection along the inside walls, HWGs may simultaneously be used as miniaturized gas cells. Thereby, the hollow core is used for confining minute sample gas volumes (i.e., usually few hundreds of microliters). With the concept of integrating straight and meandered waveguide channels into solid substrates, a new generation of HWGs has been pioneered by Mizaikoff and his research team termed substrate-integrated hollow waveguides (iHWGs) [9–14], which provide substantially improved mechanical robustness along with high optical efficiency at yet compact dimensions.

Given the almost unlimited variety of potential waveguide designs for such modular iHWGs, the achievable optical path, and associated with that the number of reflections at the inside walls of the iHWG remains limited by the attenuation of the waveguide due to reflection losses, surface roughness, etc [9]. However, also conventional HWGs require extended physical lengths (i.e., up to several meters) for achieving the demanded optical path length, yet, due to their inherent flexibility with significantly less mechanical stability compared to iHWGs. Furthermore, using extended segments of conventional HWGs inherently limits their integration into compact sensing systems. While it has been shown that fiberoptic HWGs may be coiled for decreasing their physical dimensions, the associated signal attenuation due to bending losses remains a limiting factor [7, 8, 15–19], as the attenuation coefficient varies following $1/R$ with R representing the bending radius.

In order to facilitate optical sensing applications simultaneously using hollow waveguide structures as both photon conduit and miniaturized gas cell, several features are demanded including (i) small dimensions, (ii) high sensitivity of the resulting sensing device, (iii) rapid response times/exchange of the sampled gas volume, (iv) potential for integration with other sensor components such as light source, optics, and detector, and (v) overall robustness (e.g., mechanical stability, mountability, stability against vibrations, etc). With the transformation of conventional HWGs into substrate-integrated HWGs (i.e., iHWGs) the majority of these criteria may be met. Therefore, it is anticipated that iHWGs constitute a breakthrough technology that will be adopted in a wide variety of gas sensing applications and deployment scenarios providing a viable alternative to conventional HWGs and multi-pass gas cells.

The present study focuses on design considerations for optimizing iHWGs serving as active transducer in mid-infrared (MIR; 3–20 μm) gas sensing applications with particular emphasis on facilitating their integration into the sample compartment of ultra-compact Fourier transform infrared (FTIR) spectrometers while maximizing the achievable analytical figures-of-merit.

The integration of iHWGs into ultra-compact FTIR enables designing sensing platforms capable of performing gas analysis with remarkable molecular discrimination and sensitivity, yet requiring only minute sample volumes (usually few hundreds of microliters), which is a particularly attractive feature for exhaled breath (EB) analysis associated with small animals such as mice. Furthermore, the potential of performing inline/online analysis and non-invasive monitoring is highly attractive for using such devices in early disease recognition, as the obtained quantitative results may be intimately related to biomarker panels [20–22] provided by a specific disease condition, as well as in intensive care (e.g., inline diagnostics within respirators and ventilators), and for therapy progression monitoring. Despite the challenges involved in optical EB analysis for effective usage of such sensing platforms in clinical environments including e.g., spectral interferences, adequate breathe sampling, etc, a promising strategy toward compact devices capable of sufficient molecular discrimination and quantification of volatile breath constituents at ppm–ppb levels is evident. Compact diagnostic platforms such as the currently developed integrated mouse breath analyzer (iMBA) have already successfully been tested for investigating exhaled mouse breath [9, 15, 23] evidencing the utility of this optical sensing concept in the field of breath diagnostics.

The present study compares the geometries and optical path lengths of straight versus meandered iHWGs for facilitating the functional integration into a shoebox-size FTIR spectrometer by maximizing the achievable analytical figures-of-merit in advanced breath analysis.

2. Experimental

The iHWGs used in the present study were fabricated into an aluminum substrate with varying nominal central channel length and/or geometries (figure 1). The integrated channels may be straight or meandered, thus defining the probed gas sample volume. Given the open architecture, the waveguide channel remains accessible for the deposition of additional coatings e.g., gas phase Au-deposition, etc, and/or additional polishing or surface treatment procedures prior to sealing the waveguide channel with an appropriate lid [9].

In order to evaluate the performance of straight and meandering iHWG channels, three different iHWGs with a substrate dimension of $75 \times 55 \times 15$ mm ($L \times W \times H$) were designed and milled into aluminum substrates. The corresponding aluminum cover plates were polished to a mirror-like finish, and equipped with sample inlet—and outlet connection ports. BaF_2 windows were used to cap-off the optical channel facilitating in- and outcoupling of IR radiation. The straight iHWG channels feature a nominal central channel length of 73.5 mm, and provide none, one, or two optical funnel structures facilitating in- and outcoupling of the IR beam, as shown in figures 1(a) and (d). The meandered iHWGs have a nominal central channel length of 261.1 and 418.7 mm for one-turn and two-turn meanders, respectively. Concerning the funnel structures integrated into straight iHWGs, the area available for IR radiation coupling was increased from nominally 4.20 mm^2 (2.10×2.00 mm) to 7.90 mm^2 (3.95×2.00 mm) and—except for the flared sections—the overall cross-section of the waveguide channel was maintained.

Measurements using the straight iHWG structures (see figures 2(a) and (b) were performed via integration into the

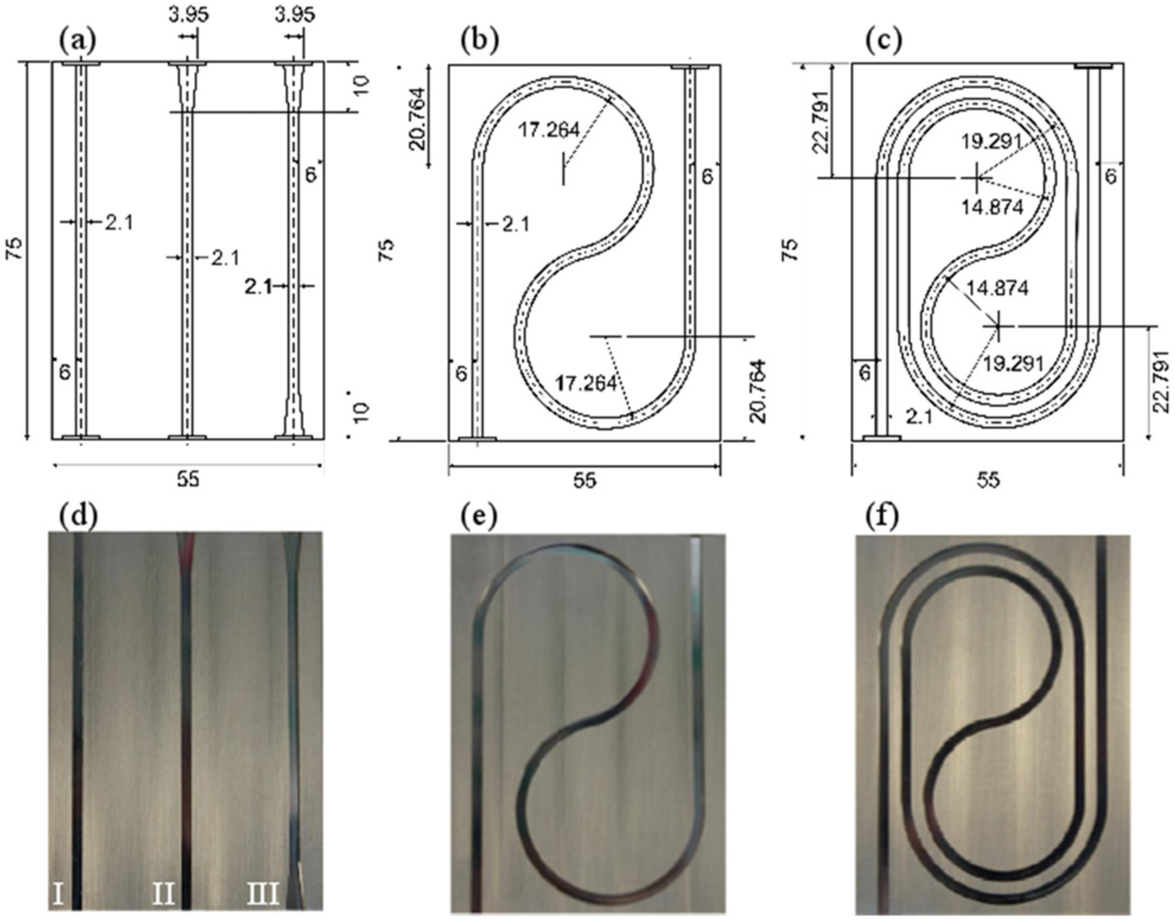

Figure 1. (a)–(c) Schematics and geometry of the iHWGs studied herein with 2.0 mm hollow core edge length. (d) Straight iHWG structures; (I): non-flared channel without optical funnel; (II): channel with incoupling funnel; (III): channel with in- and outcoupling funnels. (e), (f) One-turn and two-turn meandering waveguide channels.

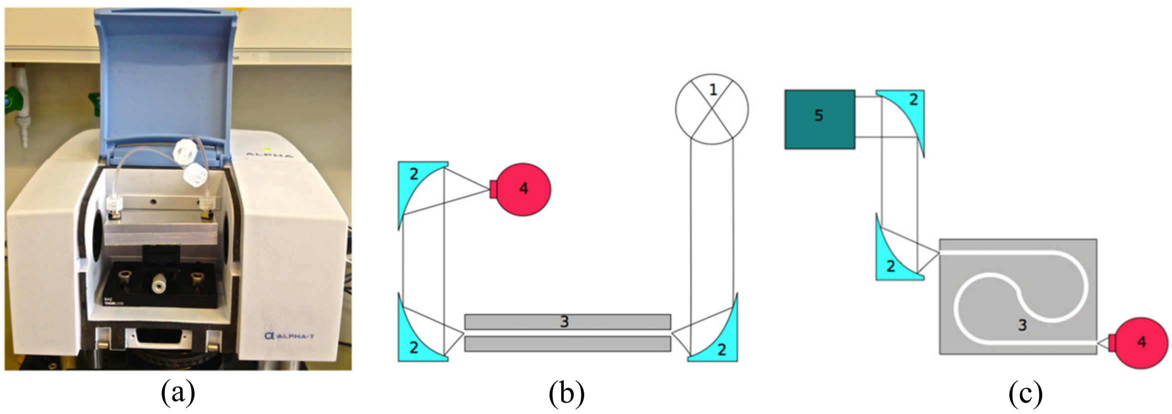

Figure 2. (a) Shoebox-size Bruker Alpha FTIR spectrometer equipped with iHWG sensing module providing straight waveguide channels with/without optical funnels. (b) Schematic of optical setup based on Bruker Alpha FTIR spectrometer with integrated iHWG. (c) Schematic of optical setup based on Bruker IRcube FTIR spectrometer with externally coupled iHWG. (1) IR source, (2) parabolic gold-coated mirrors, (3) iHWG, (4) detector and (5) FTIR spectrometer.

sample compartment of a shoebox-sized Bruker Alpha FTIR spectrometer (Bruker Alpha; Bruker Optik GmbH, Ettlingen, Germany) using the internal deuterated triglycinesulfate detector. The meandered iHWGs (figure 2(c)) were coupled to a Bruker IRcube FTIR spectrometer (IR cube, Bruker Optics, Ettlingen, Germany) equipped with a liquid nitrogen cooled mercury-cadmium-telluride (MCT) detector (FTIR-16-2.00; Infrared Associates, Stuart/FL, USA). The iHWG was coupled to the spectrometer as shown in figure 2(c) using off-axis parabolic mirrors with a focal length of 2″ (Thorlabs, Germany).

One of the main objectives of the present study was to develop an optimization procedure for sensing systems geared toward online measurements in small animal intensive care stations (e.g., mouse intensive care units) facilitated by integration of the iHWG into a shoebox-size FTIR. Given the limited space within the sample compartment of the Bruker Alpha FTIR spectrometer (see figure 2(a)), only straight-line iHWGs may be integrated. Consequently, meandering iHWGs were studied via external coupling to Bruker IRcube FTIR spectrometer. In order to ensure comparability of the obtained results albeit using different spectrometers and detectors, the obtained IR spectra were normalized and the signal-to-noise ratio (SNR) was used as a dimension-less performance parameter.

All collected IR spectra were recorded in the spectral range of $4000-650\ cm^{-1}$ at a spectral resolution of $4\ cm^{-1}$ averaging 200 spectra per measurement. The OPUS 6.5 software package (Bruker Optics, Ettlingen, Germany) was used for data acquisition and for peak area integration. Prior to the SNR calculation, the 0.1% butane spectra were normalized using the offset correction function.

The gases used herein, i.e., butane and nitrogen were purchased from MTI Industriegase AG (Neu-Ulm, Germany) and were of 3.5 and 5.0 grade.

3. Results and discussion

The channel geometry and the surface roughness are the determinant factors affecting the analytical performance of the iHWGs by limiting its energy throughput. These factors may be modeled via appropriate ray-trace techniques, thereby enabling a first theoretical optimization of different substrate materials, surface coatings (e.g., gold), and the geometry and format of the waveguide channels while facilitating integration of the substrate into an ultra-compact FTIR spectrometer. While the inner waveguide walls are polished to a surface roughness of approximately 200 nm, reflection losses still occur. In turn, this may be compensated extending the absorption path length by extending the waveguide channel length. Evidently, an optimum balancing reflection losses versus an extended absorption path length has to be determined. However, besides the theoretical optimization providing first design guidelines for iHWG fabrication, it is crucial to experimentally determine the analytical performance of the obtained structures for analyzing the practical utility of such devices in gas sensing scenarios.

In order to evaluate the analytical performance of various channel geometries (straight versus meandering) and nominal channel/absorption path lengths (i.e., 73.5–418.7 mm), a calibration function was established for each iHWG using the peak area extending from 3050 to 2800 and 1525 to $1350\ cm^{-1}$ corresponding to the band I and II of butane versus the butane concentration ranging from 0 to 1% in nitrogen. For each concentration, five replicate measurements were averaged demonstrating excellent repeatability of the gas sensing set-up (see figure 3).

Figure 3(a) compares the effect of using funneling structures for assisting efficient coupling of IR radiation into straight iHWGs. Comparing the slopes of the obtained calibration curves, the steepest slope, i.e., the highest sensitivity was obtained for a straight waveguide channel (structure II) providing a funneling structure for efficiently coupling IR radiation into the waveguide. This also reflects in the obtained SNR, as summarized in table 1 calculated and compared for the concentration of 0.1% of butane (see figures 3(a) and (b) for exemplary spectra). As expected, an optical funnel at the radiation inlet of the straight waveguide facilitates more efficient coupling of IR radiation into the iHWG; in contrast, the addition of an optical funnel at the outlet of the waveguide channel (structure III) results in reduced sensitivity, as radiation emanating at the distal end is apparently further diverged, thereby reducing coupling efficiency to the detector via the parabolic mirrors (figure 2(a)—component (2)). As expected, using a straight waveguide structure without optical funnels provides a performance in between these two concepts. It should be noted that the studies performed for comparing the three straight iHWGs were obtained using precisely the same in- and outcoupling conditions, i.e., mirror alignment, by linearly moving the iHWG substrate for addressing each of the three waveguide channels aligned in parallel within the substrate (see figure 1(d). The obtained results and system characteristics are summarized in table 1.

Analogously, the meandered iHWGs were evaluated for analyzing the effect the waveguide channel length (figure 1). A linear response can be observed for both iHWGs, however limited to concentrations <0.5% and <0.25% butane for one-turn and two-turn iHWGs, respectively, as for higher concentrations a nonlinear response of the sensor was observed. The obtained limits of detection (LOD) are summarized in table 1. From the application point of view (i.e., breath analysis), the obtained LODs are comparatively high, in particular when considering the required sensitivity for breath VOC analysis demanding for ppb ($\mu g\ L^{-1}$) to ppt ($ng\ L^{-1}$) level detection. However, suitable preconcentration strategies may readily be implemented prior to IR measurements—and similar to other breath diagnostic techniques—as recently shown for isoprene, a biomarker relevant for metabolic syndrome [24], and as previously demonstrated for analyzing benzene, toluene, and the xylenes in environmental atmospheric samples [18]. Notwithstanding, it should be noted that while improving the achievable LOD, any preconcentration strategy may (a) adversely affect the temporal resolution of the obtained analytical signal, and (b) may require significantly larger sample volumes guided through the sorbent material. In terms of dimensions, first prototypes of miniaturized thermal preconcentrators with a device footprint similar to the iHWG substrates shown herein (75×55 mm) are currently developed and tested for enriching volatile constituents from breath, and will be reported in the near future.

Regarding a suitable length of the integrated waveguide channel, this may be selected in accordance with the sensitivity demanded by a selected application, and is only limited by reflective attenuation losses. Evidently, depending on a

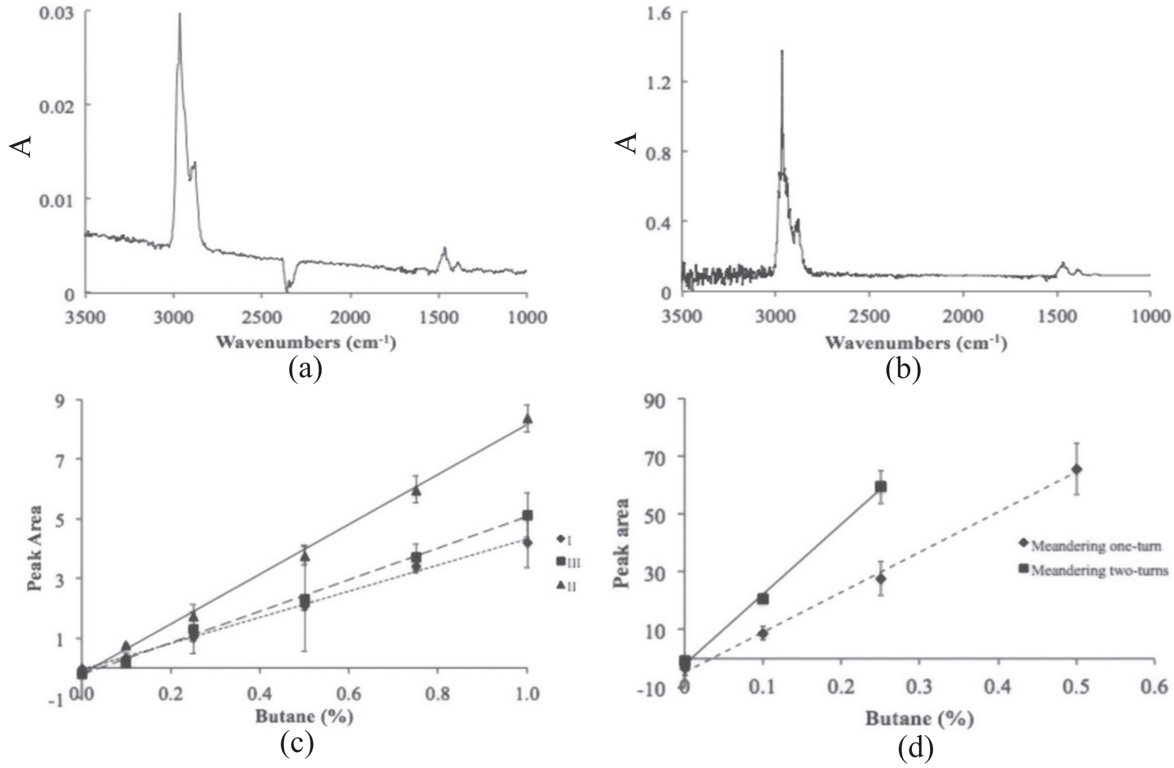

Figure 3. (a) Typical normalized spectra (0.25% butane in N_2) recorded using a straight-line iHWG with inlet radiation coupling funnel. (b) 0.25% butane in N_2 spectra recorded with IRcube for meandering two-turn iHWG. (c) Comparison of butane calibration functions within the three channels straight-line iHWGs recorded with Bruker Alpha FTIR (I, II and III refer to the structures shown in figure 1(d). (d) Comparison of butane calibration functions using meandering iHWG (one-turn and two-turns) recorded with IRcube FTIR spectrometer (for iHWG structures see figures 1(e), (f).

Table 1. Main physical and analytical parameters associated with the analyzed iHWGs.

Parameters of iHWGs	Channel geometry length (mm)/internal volume (mL^3)	Area for radiation coupling in/out (mm^2)	SNR RMS[a] (0.1% butane)	LOD (%)
Straight-line non-flared	73.5/0.31	4.20/4.20	2.73	0.42
Straight-line inlet funnel	73.5/0.33	7.90/4.20	4.98	0.13
Straight-line in- and outlet funnel	73.5/0.35	7.90/7.90	4.36	0.23
Meandered one-turn	261.1/1.10	4.20/4.20	3.36	4.10
Meandered two-turns	418.7/1.76	4.20/4.20	6.49	0.97

[a] SNR rms (root mean square) ⩾ square root of the sum resulting from the squared deviations for all data points within the selected range divided by the number of data points (selected range: 3500–825 cm^{-1})

variety of optical parameters including the radiation incoupling angle, the waveguide geometry (i.e., number of meanders, radius of curvature), dimensional constraints, etc an optimum exists for any given sensor configuration such that the increased of the absorption path length (i.e., of the waveguide channel) is outweighed by the attenuation of the radiation reflected along the light pipe. Hence, while meandered iHWGs provide an increased waveguide channel length, and therefore, an increased absorption path length for IR photons interacting with gas phase molecules, attenuation losses—which in turn depend on the coupling conditions from either a mono or polychromatic IR light source—may adversely compensate this effect. Consequently, the channel length has to be individually optimized for each sensor configuration. In the present studies, straight-line iHWGs were limited to an overall substrate dimension of 75 mm accommodating the available space within the sample compartment of the Bruker Alpha FTIR spectrometer.

4. Conclusions

We described studies on optimizing the geometry a variety of iHWG structures coupled to a FTIR spectrometer aiming at evaluating their individual analytical features. The obtained results confirm that in terms of the achievable analytical figures-of-merit, the proposed setup could indeed be applied for real-time monitoring of breath constituents in iMBA, which is specifically tailored for the rather small breath volumes (i.e., few hundreds of microliters) available during studies on exhaled mouse breath. In order to further increase the applicability, additional sample preparation steps including e.g., selective analyte preconcentration, photoreactions, etc may be integrated in-line into the gas sensing system [24].

In general, the facile integration of FTIR spectrometers with iHWGs provides a useful fundamental tool for advanced EB diagnostics, and may readily combine with complementary analytical techniques providing further insight on the molecular breath composition. The reduced dimensional format of the iHWG and the rather low sample volume required for analysis opens the possibility of monitoring a variety of volatile organic constituents in e.g., small animal intensive care scenarios where the available sample volume is limited to few hundreds of microliters such as in mouse breath diagnostics.

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