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A portable sensor system for the detection of human volatile compounds against transnational crime

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ABSTRACT

Human smuggling accounts for a significant part of transnational organized crime, creating a growing threat to national and international security and putting at risk the health and lives of the people being smuggled. Early detection and interception of human beings hidden in containers or trucks are therefore of considerable importance, especially at key transportation hubs, such as at international borders and harbors. The major challenge is to provide fast inspection procedures without needing to open sealed trucks and containers. The detection of trace key volatile organic compounds, which includes aldehydes and ketones, emitted by humans can be used to rapidly determine human presence, requiring only several ml of air to be taken from inside a container. In this paper, we describe a prototype portable device for the rapid detection of hidden or entrapped people, employing a combined ion mobility spectrometer and sensor array system for obtaining a volatile signature of human presence. The detection limits of this combined analytical device are sufficiently low for use in sensing ketones and aldehydes being emitted by humans in closed containers. For easy handling by security personnel, a classification algorithm is applied that provides a simple YES or NO decision. With a training dataset of more than 1000 measurements, the algorithm achieved an area under curve of 0.9 for untrained scenarios. The field measurements show that two people need to stay in a car for between 20 and 30 minutes in order for the emitted trace volatile organic compounds to reach concentrations high enough for reliable detection with our analytical device.

1. Introduction

The illegal movement of people across borders affects many countries, most notably the United States and the European Union. In recent years the trafficking or smuggling of people to Europe has reached epidemic proportions [1]. This not only puts a major strain on European resources, but endangers the health and lives of the people being trafficked or smuggled. Criminal networks promote migrant smuggling, offering transportation in trucks, containers and specially reconstructed vehicles, frequently at very high prices. Often the people being transported suffer inhumane conditions, being given barely any food or drink and receive little or no information about their ultimate location.

Owing to limited manpower and the lack of reliable, cheap and easy to operate search and rescue devices, security personnel are unable to cope with the high influx of people illegally entering a country. In this context, the rapid detection, and hence interception, of smuggled or trafficked people is important, not only in the interest of saving them from life-threatening situations, but also for protecting borders.

At border controls with high rates of traffic, the time available for inspection plays a critical role, because thousands of trucks and containers may cross a border every day. Therefore, any disruptions caused by security measures need to be limited. For a detection device to be applicable, a container needs to be inspected within 2–3 minutes, preferably without opening its doors or breaking any customs seals.

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1.1. Tracking systems

Many search and rescue teams increasingly rely on specialized detection systems. In Europe, search operations are currently conducted primarily with the help of x-ray scanners, CO_2 sensors and infrared heat cameras. These technologies have limitations; infrared cameras are not effective where there are only very small temperature differences between the body and the surroundings, which often occurs in the summer when temperatures around 35 °C or higher are reached, and are limited to use with trucks that have tarpaulins; CO_2 sensors have high false-positive rates, because of the elevated CO_2 emissions from organic materials such as vegetables, fruits, wood, etc.; and x ray scanners entail health risks from the ionizing effects of the radiation and hence are only occasionally used.

Dogs are still the preferred choice for quick detection of entrapped people [2], because they can rapidly track the human scent, but they have limitations. For example, dogs tire after about of intensive search. Moreover, they can become stressed and frustrated if they are unsuccessful [3]. Another issue is the significant cost involved in their training and upkeep, in addition to the cost for a trained handler. Therefore, for all these reasons there is a demand for the development of sensitive, inexpensive, user-friendly, safe and portable detectors that can help rapidly detect hidden people, either in addition to, or in place of, search and rescue dogs, that basically imitate dogs by sniffing for the characteristic scent of a human being. One approach is the real-time chemical analysis of air in small-enclosed spaces, which can give concrete indications of the presence of people. An instrumental human scent detector has the potential to significantly improve the success rate for detecting hidden people.

1.2. Human scent – emission of volatile organic compounds (VOCs)

The human body constantly releases hundreds of trace VOCs through breath, sweat, and skin [4–6], offering a continuous source of biomarkers. Volatile substances emitted from blood, urine [7] and feces [8–10] can also provide characteristic volatiles of human presence, but these are transient and therefore their emission profile rapidly changes with time. The combination of these volatiles produces a human chemical signature that can be detected with sensitive analytical chemical techniques [11].

A complicating factor that affects the use of human volatile emissions, independent of where they are coming from, is the complex background chemical matrix, which can be very diverse depending on the environment people are being kept or trapped in. The environment releases volatiles into the surrounding atmosphere that can confound the volatiles emitted by humans. Thus, it is crucial to select volatile markers that provide, as far as possible, a unique signature of human presence, which is distinct from that associated with goods in a container or from packaging. Using this criterion, the combination of CO_2 and ten trace volatile metabolites, based on the results by Mochalski [11] (ammonia, acetone, 6-methyl-5-hepten-2-one, isoprene, n-propanol, n-hexanal, n-heptanal, n-octanal, n-nonanal, and acetic acid), have been selected as being sufficiently characteristic for determining as to whether a human being is present or not with a high level of confidence.

The detection of a volatile signature for use in locating people opens up new possibilities for different search and rescue scenarios, i.e. not just those associated with illegal immigrants. For example, it includes the detection of people entrapped in a collapsed building following a natural disaster, such as an earthquake, or as a result of a bomb attack.

1.3. Portable VOC detectors

Owing to the fact that the concentrations of many human VOCs of interest are at trace levels, typically parts per billion by volume (ppb_v), their detection places great demands on analytical technologies. Furthermore, while laboratory-based state-of-the-art analytical instrumentation, such as Proton Transfer Reaction Mass Spectrometers [12,13] and Selected Ion Flow Tube Mass Spectrometers [14] can detect VOCs at such low concentrations in real-time [15], these instruments are complex and not portable. Consequently, they cannot be easily used in the field.

Analytical devices that can be carried by emergency personal in a backpack or as hand-held devices are important for on-site application. Keeping this relevant aspect in mind, gas sensors and ion mobility spectrometers (IMS) have considerable potential for use in search scenarios owing to their small size, low costs, ease of use and fast response time. Here we present details on a portable device we have developed which combines CO_2 and aldehyde sensors an ion mobility type device for the detection of key VOCs, to produce a key signature that is characteristic of humans being present.

1.3.1. Gas Sensors

Numerous companies manufacture gas sensor systems, often consisting of several sensors for multiple compound detection for the purpose of monitoring indoor air quality. The idea to use them for human scent detection is relatively new. However, the detection of human scent in complex chemical environments makes the development of a commercial sensor product challenging because of the limited selectivity involved.

There are many different types of gas sensors, e.g. optical sensors [16], semiconducting metal oxide sensors [17], and multi-walled carbon nanotubes [18], all of which show good sensitivity and reasonable selectivity for selected compounds or compound groups especially in the higher ppb-ppm range. Concerning the emitted compounds by humans, commercial available sensors are particularly effective for the detection of compounds that appear at high levels such as CO_2, acetone, isoprene and ammonia [19]. Regarding the aldehydes released by humans, detection in the low ppb-range is required. This limits the number of sensors that are commercially available. However, there are promising results associated with the detection of formaldehyde using biochemical gas sensor [20] or applying fluorescent probes based on aggregation-induced emission [21].

A commercially available electrochemical sensor reported first by Obermeier et al. [22] enables the detection of aldehydes down to the lower ppb_v levels. Its basic mechanism deals with oxidation of the aldehydes to the corresponding acids. For this purpose, it measures the oxygen consumption electrochemically. Aldehydes are oxidized at the anode, and the electrons produced by the oxidation of aldehydes are consumed at the cathode. The sensor is a typical amperometric gas sensor. The model used is this study employs an integrated circuit that converts the measured sensor current (proportional to analyte concentration) to a voltage output, which is recorded by the system.

1.3.2. Aspiratory Ion Mobility Spectrometry (aIMS)

IMS has found its greatest use in homeland security applications, primarily for the detection of chemical warfare agents and explosives [23]. However, this technology has also proven itself useful in several civilian applications, including food quality control, medical science, and industrial processes [24]. Advantages of the IMS technique with regard to the detection of VOCs emitted by humans are its robustness, portability, ease of use, high sensitivity (ng/L for ketones and amines) and rapid (seconds) and direct (without sample preparation) measurements.

A compact type of IMS, the so-called aspiratory ion mobility spectrometer (aIMS) is a promising analytical device for use in urban search and rescue (USaR) as well as chemical, biological, radiological and nuclear (CBRN) scenarios. During operation, the aIMS air is pumped continuously into the sensor cell, where the air molecules are ionized by a ^{241}AM ion source. Ion-molecule reactions with trace VOCs produce signature product ions that are spatially separated according to their mobilities (determined by the mass, charge and collisional cross-section of the ions, and the type, temperature and pressure of the buffer gas).

Product ions are detected as a current pulse. The higher the mobility of an ion, the earlier it collides with a detection electrode. To operate in either positive or negative ion mode, the polarity of the electric field is reversed in cycles typically at a frequency of 1 Hz. A histogram of ion current at each electrode is used to provide a spectrum that provides a chemical fingerprint of human presence, without identifying the individual volatile compounds. Given that the gas composition of human emission samples and the surrounding air is extremely complex, it is challenging to unambiguously match VOCs to humans. Hence, the use of chemical fingerprints rather than trying to identify individual volatiles is attractive. That is the principle adopted by us in the portable locator we have developed and are describing here. aIMS signal values containing 8 channels in positive and 8 channels in negative polarity were measured in pA then converted and read out in Volts.

2. Experimental

2.1. Set-up of the human tracking system

The portable backpack human odor tracking system we have developed combines a state-of-the-art aIMS (ChemproDM, Environics Oy, Finland), a dual channel carbon dioxide sensor module (Telaire, range: ppm, Amphenol Advanced Sensors, Pforzheim, Germany), and an aldehyde sensor (IT Dr. Gambert GmbH, Wismar, Germany).

The housing, containing the key components of the aIMS, is water-resistant and can be combined with a backpack carry system (Fig. 1a-c). The gas sample is collected from the end of a thin (1/8 inch) inert flexible tube, which makes it convenient for inserting through small holes (e.g. through a plughole for condensed water in sealed containers). Using a specially designed sampling system, including inert tubing, dust separator and pump, the gas sample is transported to an inner gas circuit containing an aldehyde detector, a CO_2 sensor and an aIMS. This ensures the continuous high airflow rate required for the aIMS (up to 1 L/min) triggered by an in-built pump. Thus, the sampling pump runs only when collecting air or flushing the system with clean outside air.

2.2. Data acquisition and agglomeration

A single-board Raspberry Pi 3B is employed as the core-processing unit to collect the data from all detectors. It runs on a Linux operating system and provides several hardware interfaces to communicate with the detectors and peripherals. Dedicated C++/QT software continuously collects data from the different sensors and stores the data in a database during acquisition intervals. Fig. 2 schematically shows the software architecture. The measurement protocol runs continuously and requests data from the aIMS device, which is connected via a RS232 interface every second. Additionally, a second interface is available for communication with other programs providing a live stream of data via the Websocket protocol. This controls data acquisition with the handheld sampler or a web interface. Possible commands include the starting and stopping of acquisition, and the enabling of a stream of live data for plotting or for diagnosis on the web interface. Additionally, the same socket is used for other data, including the fusion of CO_2 or aldehyde values with the measured aIMS data.

During an acquisition interval, data are stored inside a MySQL database. Besides the raw sensor values, additional information, such as a timestamp, comments and type of measurement, are saved to the database. All data acquired in a measurement sequence can later be accessed foror as part of the analysis routines. When using an external database, different programs can access the same data simultaneously, thus allowing a central and shared database server to be used by multiple measurement units. However, in the present prototype the database runs only on the Raspberry Pi.

2.3. Test gas preparations

Liquid standards of selected VOCs, namely acetone, 6-methyl-5-hepten-2-one, isoprene, n-propanol, n-hexanal, n-heptanal, n-octanal, and n-nonanal were purchased from either Sigma, Sigma-Aldrich or Fluka, with stated purities greater than. Test gases of these compounds were prepared after evaporation of the liquid standards in the concentration range of ppb as described in [5] and measured with the aIMS and aldehyde sensor.

Using the gas standards, the limits of detection were calculated with

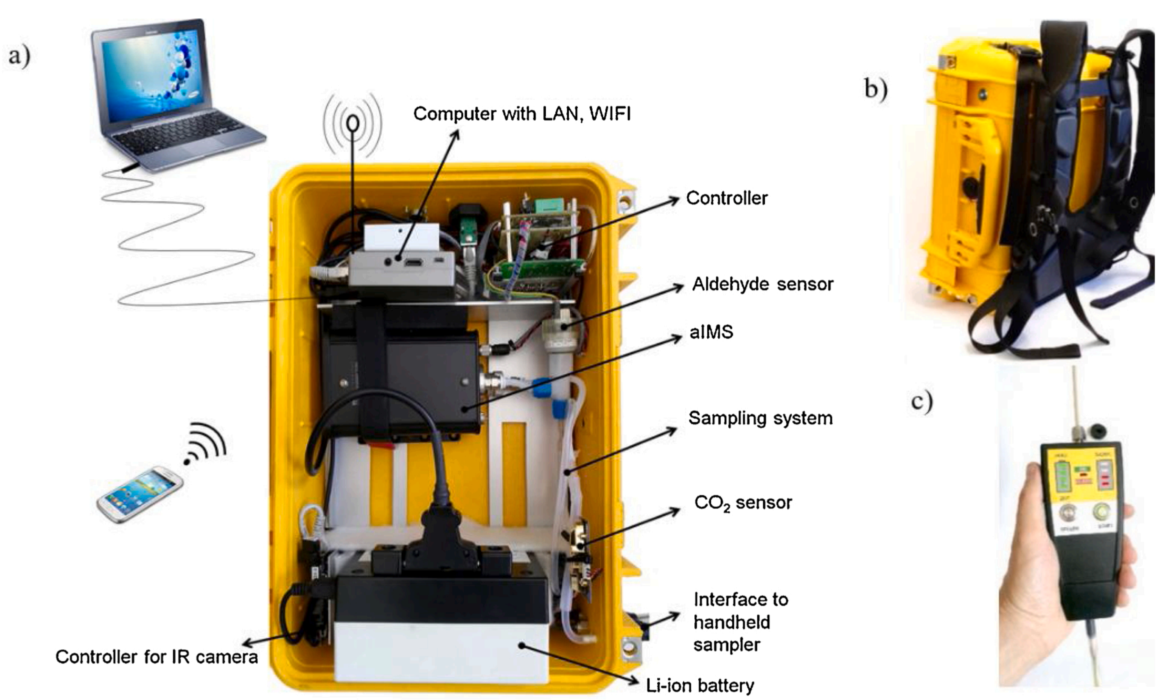

Fig. 1. The portable backpack human odor tracking system showing a) its set-up, b) the back-pack carry system, and c) the handheld sampler.

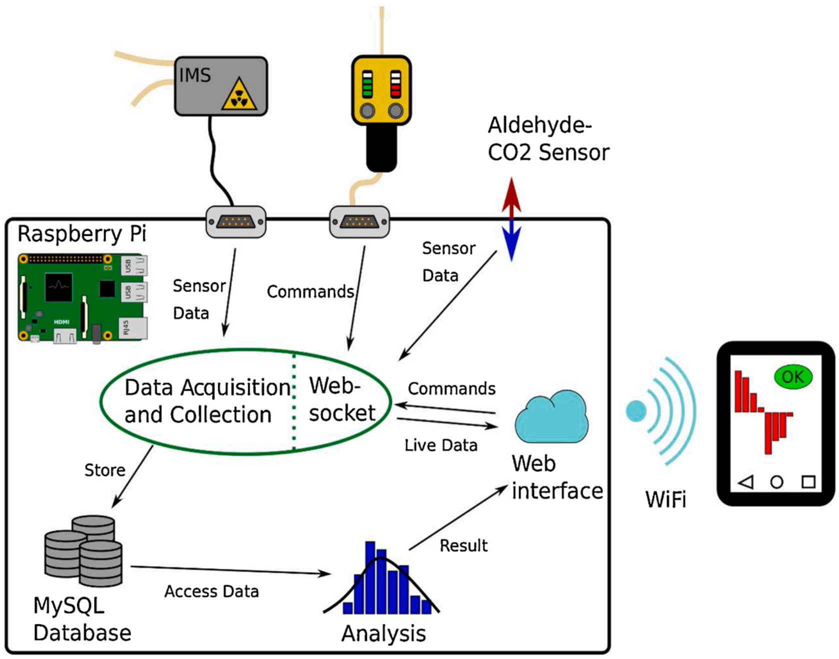

Fig. 2. Schematic of the hardware, software and interfaces used for data acquisition, visualisation and analysis.

the signal-to-noise ratio of 3:1 in the case of the aldehyde sensor and were based on visual evaluation (minimum level at which an analyte can be reliably detected) for the aIMS.

2.4. Sampling protocol for field tests

To mimic field situations as well as possible, the following requirements were set for the training localities: closed rooms were selected with a maximum volume of 20 m^3 and minimal ventilation. One to four people were placed in the room for at least ten minutes.

Before starting the measurements, the room was kept empty of people for at least 20 minutes in order to record background VOC levels, with these levels being monitored 15 times alternatively for the inside and the outside air. Subsequently, between one and four people entered the room. After ten minutes, 30 measurements of the inside and the outside air were undertaken. The total measurement time was about one hour. This protocol was repeated several times - also on the same day – with appropriate ventilation of the room between each series of measurements.

2.5. Signal quality control in the field

Determination of a sensor's sensitivity directly in the field is of utmost importance to ensure correct measurement of volatile signatures. For this purpose, a calibration system using a permeation device was constructed to permit regular monitoring of the instrument's functionality and sensitivity.

The permeation tube consists of a polyether ether ketone polymer (PEEK) holder sealed around a perfluoroalkoxy alkane polymer (PFA) tube (external diameter 1/8th inch, wall thickness 1 mm) filled with a liquid compound (Fig. 3). Molecules of the liquid diffuse through the wall of the PFA tube and are carried by the gas (air) streaming through the tubing. A constant gas flow and permeation rate of the contained substance at a constant temperature results in a stable compound concentration.

By switching the permeation tube to the inlet of the sensory system and starting sample collection, outside air is pumped through the tube resulting in an elevated concentration of the selected compound in the sample. The measured signal provided the information needed to determine the sensitivity of the device.

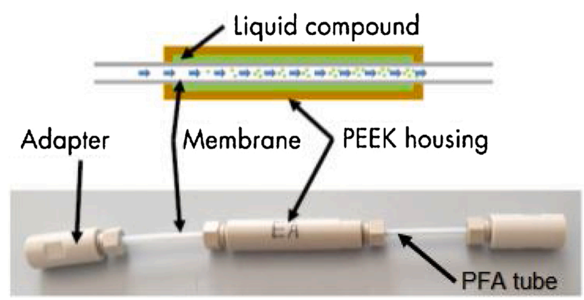

Fig. 3. Set-up of permeation tube for signal quality control.

Ideally, the concentration of a selected substance in the sample is between the quantification limit and the saturation level, which depends on the sensitivity and the linear range of the analytical system for a given compound. Thus, for aIMS the concentration should be between 50 and 200 ppb, which with the current set-up using a gas flow of 250 ml/min for ethyl acrylate is reached in the temperature range 15–35 °C. Owing to the linear relationship between the permeation rate and the temperature, the anticipated signal can be calculated depending on the outside field temperature. However, for a quick functionality test (sensor is working YES/NO) any concentration in the range is suitable, and thus the temperature dependency can be ignored.

It must be pointed out that if the permeation tube is stored without flow, it takes up to 30 minutes of flushing to achieve an equilibrium. Thus, for storage, the permeation tube should remain connected to the gas outlet of the sampling system to maintain the equilibrated state. This permits the function tests to be conducted immediately when the permeation tube is switched from the outlet to the inlet.

2.6. Data processing algorithms

For machine learning, a „Random Forest "method was adopted [25]. The following parameters were employed from the measured data from 16 aIMS channels (eight for positive mode and the other eight for negative mode), a CO_2 sensor, an aldehyde sensor, and pressure, humidity and temperature sensors. Half of the acquired data were randomly allotted to a training set and half to a validation set. A random-forest classifier was constructed using the training set, which was tested on the validation set (thus on data not used for training purposes). Machine learning was implemented using the software H2O Flow [26]. The optimised setting was determined according to Cook [27]. The generated classifiers were exported as objects for the program language JAVA (keyword POJO), which were delivered to the sensor system as a command line program that employs data in the format JSON (ISO/IEC 21778:2017) as input and output.

3. Results and Discussion

3.1. Detection limits

Test gases of the selected volatiles were measured separately with the aldehyde sensor and with aIMS. The detection limits for the compounds are displayed in Table 1. As expected, aIMS shows less sensitivity for the aldehydes, but greater sensitivity for the ketones, such as acetone and 6-methyl-5-hepten-2-one. From previous experiments that determined the volatile emission profile of humans in a body plethysmograph chamber [11], it is known that the detection of aldehydes at low ppb levels is important in order to capture the human scent, which can be done with the aldehyde sensor. The selectivity of the aldehyde sensor is controlled by the porous polytetrafluoroethylene membrane. Interestingly, besides the aldehydes, isoprene, which is a hydrocarbon, is also detected by the aldehyde sensor in low concentrations. We are not able to explain this cross-sensitivity to another compound class, but this provides an analytical advantage, since isoprene is also a key biomarker of people.

The decreased sensitivity for aldehydes with aIMS results from the high humidity altering the ion-chemistry. If the moisture content of the sample is separated, e.g. with a chromatographic column, IMS devices reach detection limits in the low ppb range for aldehydes [11]. However, trapping out water is not an option for breath measurements, because it would also result in a loss of trace level compounds and further complicates a field device. We cannot rule out the cross sensitivity for other compounds e.g. unsaturated hydrocarbons associated with volatiles in human breath.

The limitations of the compact aIMS with regards to detecting aldehydes in the required low concentration range is overcome by using a device that combines the aIMS with an aldehyde sensor.

3.2. Field test results

Detection of CO_2 in conjunction with VOCs is important, because it further enhances the analytical capabilities of the instrument. Given its high concentrations in human breath, CO_2 is easily detected and can therefore be used as an initial possible sign of life. If CO_2 is detected, the more sophisticated aIMS detector and the aldehyde sensor can then be used to search for a signature reflecting human-specific VOCs. Hence, the combination of CO_2 and volatile detectors (aldehyde sensor and aIMS) provides a highly specific analytical tool, with which first responders or security personnel can detect the presence of trapped or hidden human beings with a high level of confidence.

Altogether 1240 measurements of outside air, 430 measurements of air inside an empty sealed interior such as a small room, container, car, transporter, and 1023 air measurements with human beings present in a closed area were acquired using the protocol described above. These measurement were used for training and the evaluation of the software algorithms for separation of people present in a closed volume (group 1) and closed volume without humans (group 2).

The result of the algorithms is a classification score ranging between 0 and 1. Note that this number does not represent a probability, but a likelihood. A higher score means a greater likelihood of a true positive (chance of a person present being correctly detected) and lower likelihood of a false negative (risk of a wrong non-detection).

Fig. 4 provides an example of readout values obtained when using the protocol described above in the field, resulting from measurements of air inside and outside a container, with two people sitting inside at two different times, with a suitable ventilation break between the two. This demonstrates the reproducibility of the measurements.

Values determined from the measurements alternating between inside and outside air show that 90 seconds is a sufficient time for all three steps, including air sampling, reproducibility measurements and rinsing out the air sample. The measurements of the surrounding air outside show classification scores that are much lower, nearly constant, as compared to those measured in the car during the whole procedure. The intensities calculated for the inside air increase in the first 10–15 minutes and then reach a more stable value.

In the field, a fast and clear analytical decision is required that alerts security personnel that hidden people might be present. Therefore, score values above the threshold is converted to a red light on the display of the handheld device, whereas score value below the threshold are converted to green light, which indicates no hidden people. This decision is made by comparing the classification number with a predetermined threshold from the training data (Fig. 4). The threshold value is based on the receiver operating characteristic (ROC) curve [28]. From the experiments we determined that threshold values ranging from 0.3 to 0.4 are sufficient, giving excellent results as seen in the almost perfect ROC curve shown in Fig. 5a (based on the data set achieved from the trained case). However, in everyday practice it may be necessary to adjust the threshold to accommodate for deviations from the trained data set. For each threshold, sensitivity (rate of positively detecting people controlling the rate of missed detections) and specificity (rate of correct no-alarm controlling the rate of false alarms) are estimated. An acceptable combination of these two values can be chosen and the corresponding threshold selected. Sensitivity can be improved only at the cost of specificity, and vice versa. This decision needs to be made strategically, namely by judging current risks and weighing the cost of false alarms and failed detection. In the training data set the classification works extremely well (Fig. 4, left), so that values from 0.3 – 0.4 deliver perfect scores (giving a value of almost 1.0) for sensitivity. However, when applied to an untrained situation (for example, Fig. 4, right) both qualities depend on the threshold sensitivity. In this example, a sensitivity and specificity of 0.8 are simultaneously achieved.

Fig. 5 (a) shows the ROC curve based on the validation data for the trained case (according to the experimental protocol described in Section 2.4 without any organic background emissions providing cofounding signals, such as from vegetables, fruits, etc.). In this scenario, the area under the curve (AUC) is greater than 0.999, which means the classification is almost perfect. For the untrained case, displayed in Fig. 6 (such as inspected trucks with any delivered goods including containers filled with vegetables), the AUC is around 0.9 with a sensitivity and specificity of 80% each. However, this result can be improved with additional measurements when following two different strategies. On the one hand, the classification number can be trained for all possible

Table 1
Detection limits for the ten selected VOCs for the aldehyde sensor and aIMS. This illustrates the need for the combination of the two analytical devices for use in the field device to detect hidden people.

VOC	Detection Limits Aldehyde Sensor	Detection Limits aIMS
Acetaldehyde	25.8 ppb	> 50 ppb
Propanol	7.0 ppb	> 100 ppb
Hexanal	4.9 ppb	> 50 ppb
Heptanal	13.8 ppb	> 50 ppb
Octanal	8.2 ppb	> 50 ppb
Nonanal	14.9 ppb	> 50 ppb
Acetone	> 500 ppb	2 ppb
Acetic acid	> 500 ppb	> 50 ppb
6-Methyl-5-hepten-2-one	5.0 ppb	1.5 ppb
Isoprene	8.5 ppb	> 200 ppb

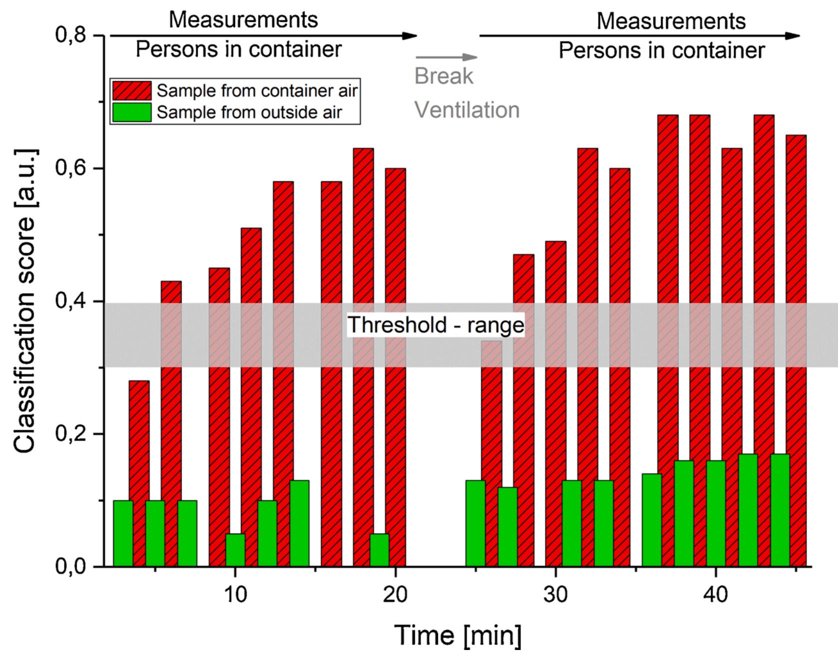

Fig. 4. Classification numbers calculated during alternating measurement of the air inside (shaded columns) and outside (unshaded columns) a container with two people sitting inside.

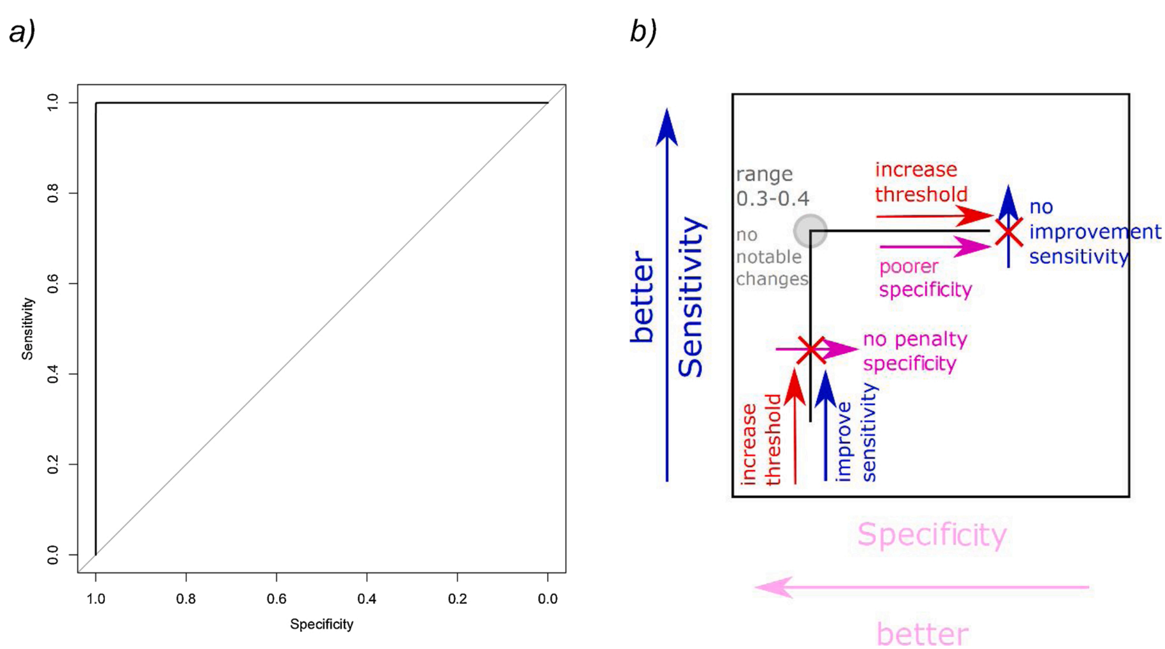

Fig. 5. (a) ROC curve for the trained case (people in a closed room or a container without organic materials transported), and (b) threshold range 0.3-0.4 is ideal for an almost perfect ROC curve (AUC~1).

transported products, which means very large numbers of measurements and unrealistic goals for the system; for instance, it should recognise the delivered wares from their smell, exclusively, or the type of container to be sampled. On the other hand, a more practicable way would be to determine a series of classifiers for typical scenarios, from which the right one can be selected at the inspection point. In practice, the working point at the ROC curve has to be selected in such a way that the costs and efforts of the inspection action are optimised. For this purpose, the ratio of positive to negative results and the ratio of false-positive to false-negative results should be considered (e.g. if large numbers of containers are to be inspected in one day, sensitivity can be lowered in order to save time and money, because the prototype human scent analyser has such a high success rate).

The work described in this paper is a result of collaboration between the Institute for Breath Research at the University of Innsbruck, the Austrian Federal Ministry of National Defense and Sports, the Austrian Federal Ministry of the Interior, Ionicon Analytik GmbH and Austrian Johanniter Unfall-Hilfe. Such an intersectoral approach provides important input into the application-oriented research and takes into account social and ethical issues. Importantly, it ensures that the developed portable device is designed according to user requirements and that it can be readily adapted for use with other detection methods currently in the field.

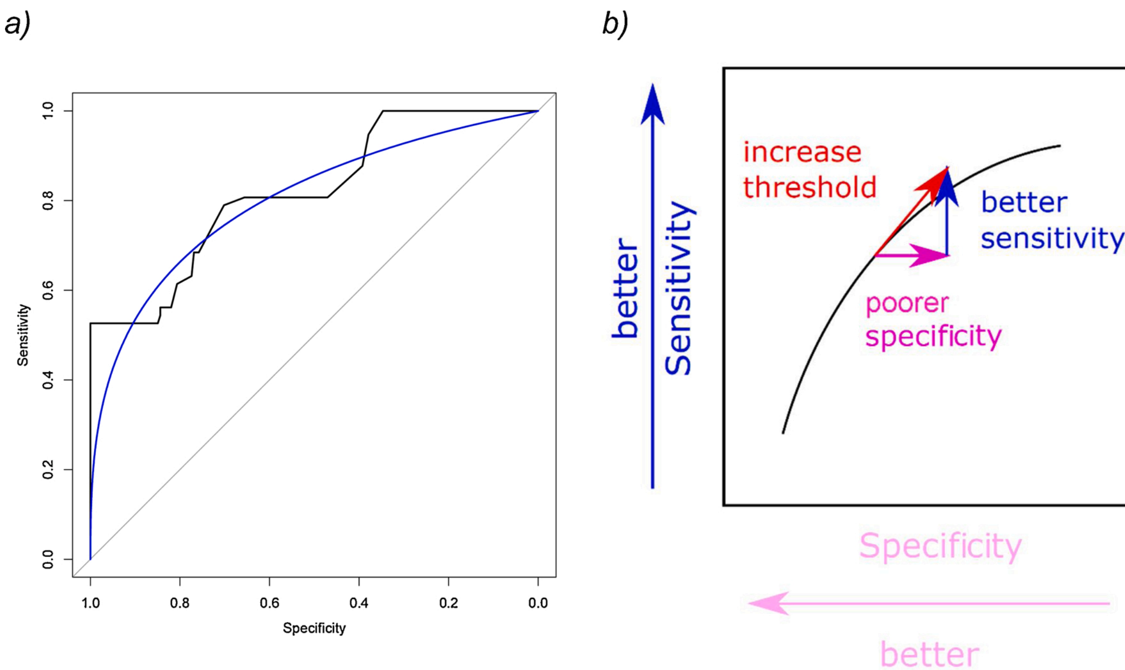

Fig. 6. (a) ROC curve for the untrained case (people in a container together with vegetables/fruits transported) and (b) expected effects for changes in the threshold. The black line is ROC from the measured actual data connected by straight lines to emphasize the curve. The smoothed line gives an expression how the underlying ROC looks like.

CRediT author statement

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J. Herbig: Optimisation of aldehyde sensor, permeation tubes
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M. Fischer: Firmware, Software, Electronic parts
M. Renzler: Firmware, Software, Electronic parts
T. Ussmueller: Firmware, Software, Electronic parts
K. Lindner: Writing
C. A. Mayhew: Writing, Conceptualization.

Declaration of Competing Interest

The authors report no declarations of interest.

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