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Food Freshness Measurements and Product Distinguishing by a Portable Electronic Nose Based on Organic Field-Effect Transistors

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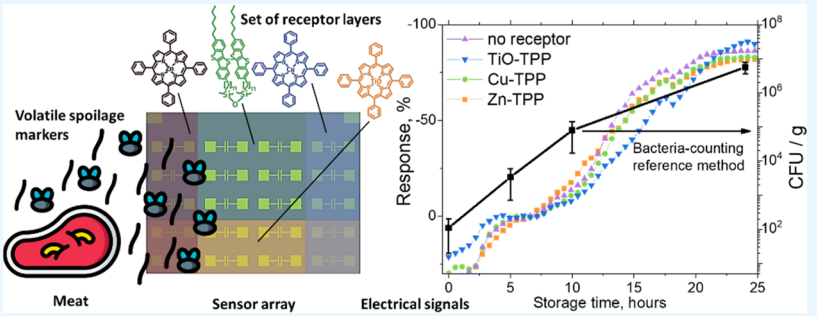

ABSTRACT: Determination of food freshness, which is the most ancient role of the human sense of smell, is still a challenge for compact and inexpensive electronic nose devices. Fast, sensitive, and reusable sensors are long-awaited in the food industry to replace slow, labor-intensive, and expensive bacteriological methods. In this work, we present microbiological verification of a novel approach to food quality monitoring and spoilage detection using an electronic nose based on organic field-effect transistors (OFETs) and its application for distinguishing products. The compact device presented is able to detect spoilage-related gases as early as at the 4×10^4 CFU g^{-1} bacteria count level, which is 2 orders of magnitude below the safe consumption threshold. Cross-selective sensor array based on OFETs with metalloporphyrin receptors were made on a single substrate using solution processing leading to a low production cost. Moreover, machine learning methods applied to the sensor array response allowed us to compare spoilage profiles and separate them by the type of food: pork, chicken, fish, or milk. The approach presented can be used to monitor food spoilage and distinguish different products with an affordable and portable device.

INTRODUCTION

Sensor arrays or electronic noses for food quality assurance and quality control (QA/QC) have been developed for decades.[1,2] Meat spoilage is accompanied by a release of various sulfur- and amine-containing compounds, carbonyls, hydrocarbons, and alcohols, in detail determined by headspace gas chromatography with mass spectrometry (GS/MS) studies.[3,4] For example, spoiling poultry is known to release sulfuric compounds, such as dimethyl disulfide, dimethyl trisulfide, phenyl sulfide, methyl thiolacetate, alklyl methyl sulfide, and 2,4,6-trimethylpyridine.[5] Ethanol, acetone, pentane, and dimethyl disulfide were also identified from chicken packages with a modified atmosphere.[6] An electronic nose capable of detecting food spoilage should have sensitivity toward some of the compounds related to off-odors and protein decomposition in the ∼100 ppb range and operational stability under a high level of relative humidity, preferably at low refrigerated temperatures.[7,8] Fast, sensitive, and reusable sensors are long-awaited in the food industry to replace slow, labor-intensive, and expensive bacteriological methods or GS/MS.[8,9] Further sensitivity enhancements would pave the way to the automated determination of early food spoilage that can be used in smart fridges, storage rooms, or even in smart packaging. This would drastically reduce unnecessary food waste, which is now up to 30% of all of the food produced,[10] and prevent many cases of food poisoning.

Colorimetric sensor arrays based on dye dots are promising for the food industry, especially in smart packaging,[3,11] but disposable components limit their implementation when continuous measurements are required. The arrays based on metal−oxide semiconductor (MOS),[12−14] quartz crystal microbalance (QCM),[15] and conductive polymer (CP)[16,17] novel nanosensors are known so far and have already been applied to a large number of sensing applications. At the same time, they are still bulky and costly compared to self-assembled

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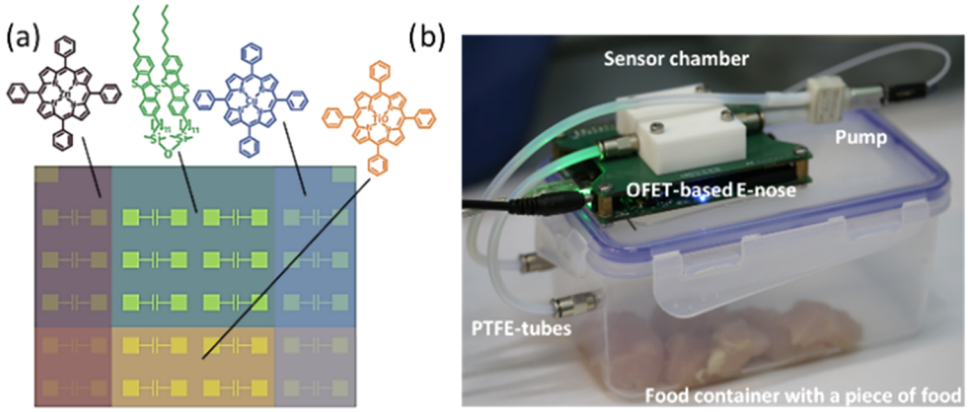

Figure 1. (a) Schematic representation of the sensor array with 20 monolayer OFETs, where different colors represent the areas covered by various receptor layers (TiO−, Cu−, and Zn−TPP) and nonmodified sensors in the substrate center with the bare D2-Und-BTBT-Hex organic semiconductor. (b) Scheme of the experiments with a food container connected to the sensor chamber via a micropump.

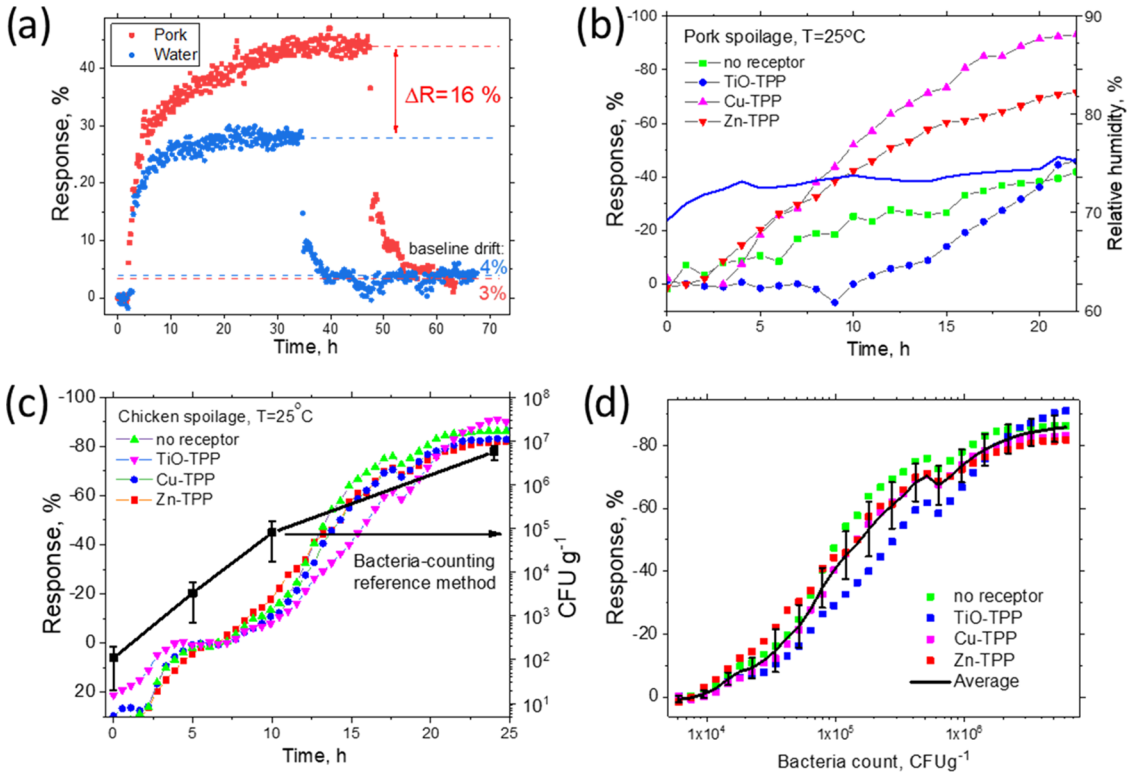

Figure 2. (a) Comparison of the nonmodified D2-Und-BTBT-Hex sensor response over distilled water (blue) and a piece of pork (red) stored in a fridge at +5 °C. (b) Response−time dependencies on spoiling pork for four sensor groups with different receptor layers and relative humidity measured with a reference sensor (plotted as the blue line and linked to the right axis). (c) Response−time dependencies on spoiling chicken for four sensor groups with different receptor layers and colony forming unit (CFU) counts (plotted as the black line and linked to the right axis). (d) Sensor response recalculated over bacteria count per gram units.

or printed organic electronic devices, often require heating, and have limited sensitivity.[12,13]

The method of meat freshness determining by a sensor printed with ink on chromatographic paper was recently described.[18] It is sensitive to water-soluble gases being the markers of meat spoilage. In particular, the authors showed sensitivity to ammonia in the range of 0.2−1000 ppm and the sensor's ability to determine the freshness of chicken or fish. The disadvantages of this method are the lack of selectivity between various water-soluble gases, which leads to the impossibility of distinguishing them, the short lifetime of such sensors, and the dependence of the response on relative air humidity.

Modern semiconductor gas sensors allow achieving the ppb-level limit of detection, while their cost is much less compared to the benchmark optical and mass-spectroscopy equipment. Recent work on inorganic semiconductors describes a selective gas sensor array capable of a particular food decomposition assessment.[19] However, protein decomposition has a complex profile that is difficult to be linked to a single responsible marker such as hydrogen sulfide or ammonia. Moreover, storing multiple food products together blurs the signal with background volatile compounds. Commercial MOS-based

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electronic noses published previously can be used for food spoilage monitoring. However, these devices are bulky and energy-consuming since they include more than ten separate sensors requiring heating.[6,20]

Organic field-effect transistors (OFET) are especially promising as gas sensors due to their high sensitivity, reusability, low power consumption, and cheap production.[21,22] However, single OFET-based sensors are sensitive to a wide variety of analytes leading to their main drawbacks—poor selectivity and environmental instability.[23−25] These reasons limit their application for food quality analysis, which often occurs at a high background relative humidity level, especially in the fridge environment. A combination of semiselective sensors in arrays can overcome poor selectivity. In this work, we describe a microbiological verification of the OFET-based electronic nose to food quality monitoring and its application for distinguishing various product spoilage profiles. This approach relies upon a whole sensor array production by self-assembly on a single substrate and simple electronics, which results in their compact size and low cost.

RESULTS AND DISCUSSION

In this work, we have used a sensor array of monolayer OFETs based on the siloxane-containing derivative of [1]benzothieno-[3,2-*b*][1]-benzothiophene organic semiconductor (D2-Und-BTBT-Hex).[24] Its sensor response selectivity was tuned with different receptor layers on a single substrate (Figure 1a). A detailed description of the sensor array and measuring device fabrication was recently published elsewhere.[27] Initially, a set of metalloporphyrin receptors (TiO−, Cu−, and Zn−TPP) was chosen to provide discriminative ability between four gases: NH_3, H_2S, ethyl mercaptan (Et-SH), and NO_2. At the same time, various amine-containing (such as methylamine, dimethylamine, and cadaverine) and sulfur-containing compounds (such as dimethyl sulfide) with comparable or higher molecular weights compared to the gases mentioned above are reported to be markers of protein decomposition and thus can be used to detect food spoilage, meaning that the same sensors can be applied to detect them.[3−5,28]

The sensors show up to 200%/ppm sensitivity to ethanethiol with a below 40 ppb LOD,[26] so they are capable of detecting spoilage byproducts. That is why the objective of this work was to determine the resolution of the food freshness sensor array proposed and to study discrimination between the spoilage profiles of different types of food.

Food Experiments Design. The electronic nose was attached to a sealed container with the product of interest placed inside to examine gases released during its spoilage. The sensor chamber was connected to the food container by PTFE tubes through a small air pump, inducing airflow (the setup is shown in Figure 1b). At first, the container was filled with distilled water and left in the fridge at 5 °C for 3 days to prove that the sensor response observed is not related to a bare humidity oscillation or sensor degradation on the timescale investigated. The evolution of the nonmodified sensor response during the reference experiment is shown in Figure 2a, blue graph. On the same timescale, the response related to the experiment with pork at +5 °C is presented (Figure 2a, red graph). It can be seen that while both sensors have responses to bare humidity, the response to pork is higher than the reference signal by an additional 16% which is related to spoilage gases released during pork storage in a closed food container. Also, it can be seen that the responses are reversible, with the values returning to the initial baseline after purging the sensor chamber with clean, dry air. The baseline level has only shifted by 4% during a 70-h-long experiment. Bacteriological studies have not shown significant CFU/g growth during the 2-day-long experiment in the fridge, which is in accordance with the literature data.[6,19] Since more than two days were required to obtain a significant sensor response and bacteria counts for each food product stored in the fridge, further experiments have been conducted at room temperature to speed up the spoilage process. Before each experiment, the food container was filled with a specific kind of food product (chicken, pork, fish, or milk) and placed at room temperature (+25 °C) for 25 h until a significant signal growth was detected. We would like to note that the extension of the presented results to the fridge environment should be carefully taken since the compound profile may vary at lower temperatures, and this aspect requires additional investigations. However, previous studies have shown that during chicken spoilage, sulfur-containing compounds, such as dimethyl sulfide, are released at different temperatures, but their concentration may vary significantly.[6,19] In the GS/MS study of chicken spoilage volatiles at 2 and 10 °C,[29] hydrogen sulfide, methyl mercaptan, dimethyl sulfide, dimethyl disulfide, methyl acetate, ethyl acetate, heptadiene, methanol, and ethanol were detected. That is why the presence of these compounds at higher spoilage temperatures was expected.

Qualitative Monitoring of Food Spoilage. The electronic nose sensor response−time dependences over the pork and chicken pieces are shown in Figure 2b,c, respectively. Each line represents the response averaged over a group of sensors with the same receptor layers: TiO−, Cu−, Zn−TPP, or the bare organic semiconductor D2-Und-BTBT-Hex. Since the sensors demonstrate sensitivity to water vapors, which are also released by the food products after the container connection, the baseline values were taken after relative humidity in the food container reached an equilibrium value of ∼75% relative humidity (RH), the values of which are shown in blue in Figure 2b and linked to the right axis. Further signal growth after this point is only related to the spoilage gases released. Figure 2b shows the signal evolution over a piece of pork. The sensor response slowly grows at a constant relative humidity and reaches 40−100% after 20 h of storage. These values are 4 to 10 times higher than the baseline drift in the range of a few days, as shown in Figure 2a. The response varies between the sensor groups with different receptors allowing to catch the odor "fingerprint" of the product under investigation, as discussed below. Similar behavior is observed for the experiments with a piece of chicken breast shown in Figure 2c. It can be described the same way as the pork but with slightly different dynamics of the specific sensor responses since a particular composition of the spoilage byproducts can vary.

To cross-validate the findings and qualitatively determine the freshness of the chicken sample, a bacteriological method was used with all details given in the Experimental Section and Supporting Information (Figures S1−S3). Colony forming units (CFU) per gram were counted at 4 periods, and the data are depicted on the right axis (black line) of Figure 2c. Starting from the values below 10 CFU g^{-1}, which confirm the initial product freshness, it reaches 10^4 CFU g^{-1} on the 5th h of storage, growing up to 10^7 CFU g^{-1} on the 24th h. The critical threshold of the bacterial concentration after which the food consumption is not considered safe is estimated as 10^7−10^9 CFU g^{-1} for most types of food.[30,31] We have approximated

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the bacteria count with an exponential growth model to recalculate the response dependence over CFU g^{-1}, as shown in Figure 2c. Considering the baseline variation of below 10%, we can confirm that the device can detect the spoilage at the 4 × 10^4 CFU g^{-1} bacteria count level, which is 3 to 5 orders of magnitude beyond the threshold values. Thus, the electronic nose described could be used to detect chicken spoilage byproducts well before it becomes unsafe to consume, which has the potential to alert the user in advance and avoid unnecessary waste. The same experiments were also conducted with fish (salmon) and milk, as discussed in the next section, where the difference between the sensor responses is analyzed with machine learning approaches to distinguish features of various product specific spoilage profiles.

Distinguishing the Type of Spoiled Food by Its Odor. To determine the type of spoiled product among the other products, the proposed device with four groups of sensors having different receptor layers and thus selectivity was used in conjunction with machine learning algorithms. The measured response values of the sensor array based on OFETs together are combined into a response vector, which can be used to reduce the sensor array response vector dimensionality with principal component analysis (PCA) and to classify it with linear discriminant analysis (LDA) or artificial neural network (ANN) algorithms.[7,8] The vectors obtained in the described experiments were compared with each other or with the training dataset. For more intuitive and clear visualization, we have used the PCA method to visualize the measurements in the 2-dimensional diagram; Figure 3 shows a diagram for the

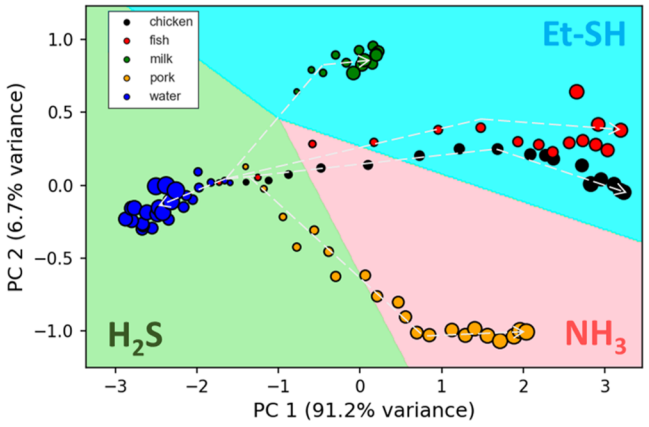

Figure 3. Principal component diagram of the food product spoilage measurements (chicken, pork, fish, milk) and water as a reference projected onto a training dataset with reducing gases (NH_3, H_2S, Et-SH). Storage time growth is shown with a circle size increase and gray arrows.

chicken, pork, milk, and fish spoilage profiles and the water signal as a reference with arrow directions and circle size showing an increase in storage time. One can see that most of the food products point in different directions, which results in the ability to differentiate their spoilage profiles by electronic nose measurements and a simple dimensionality reduction algorithm. Similar chicken and fish spoilage profiles determined by the electronic nose can signal similar gases released while proving this requires additional measurements. Three-dimensional (3D) principal component space demonstrated better product separability (see Supporting Information, Figure S4). The LDA diagram applied to the same dataset shown in Figure S5 did not provide additional knowledge in this particular case but could lead to better device training with more food types studied.

Additionally, the electronic nose device was pretrained with simple gases as published elsewhere[25] to further analyze the composition of the gases released by different products during spoilage based on the vector comparison method described above. The smallest distance to one of the training dataset vectors determines the type of spoiled product and can be applied to identify the sample. We have used the measurements collected in our previous work[25] with NH_3 and H_2S at concentrations of 0.1−1.5 ppm and Et-SH at concentrations of 0.04−0.2 ppm as a training dataset for principal component analysis transformation and marked corresponding regions by the logistic regression classifier (see the Supporting Information, Figure S6). In Figure 3, the food spoilage measurements are projected onto this marked space, where filled regions correspond to the training dataset with NH_3, H_2S, and Et-SH, and the circles are related to different food species with the arrows and circle size showing the storage time growth. The chicken, fish, and milk spoilage-related points fall deep into the Et-SH region with the storage time increase, while the pork spoilage-related points lie in the NH_3 region. All of the points related to water are shifted from the initial point due to non-zero sensor sensitivity but lie in a tiny region corresponding to a constant gas composition. At the same time, the storage of pork, chicken, and fish are followed by a signal growth with a clear direction. These preliminary results figure out the possibility to appropriately pretrain the electronic nose for the product task, leading to more accurate spoilage gas composition determination. For better device training, analytes such as dimethylamine, dimethyl sulfide, and cadaverine could also be included.[7] Based on the data obtained, the described device can detect meat spoilage byproducts attributed to the actual bacteria count for the chicken samples at 25 °C and can be used to monitor spoilage and detect it well before unsafe CFU g^{-1} levels are achieved. It can be used to assess and monitor the quality of raw food on the production lines of the food industry, control spoilage of meat, fish, and dairy products, and recommend the user to cook a particular product if signs of its imminent deterioration appear before the expiration date.

Additionally, we suppose that the presented device could potentially be applied to monitor and detect spoilage in a fridge environment, which requires additional long-run studies at lower temperatures. If substantial datasets at varied temperatures could be obtained, calibrating the sensor readings to an actual CFU g^{-1} would be possible while also requiring further studies. As a result, the device user will be informed about the freshness of the product and have a forecast for its shelf life for safe consumption, irrespective of the formal expiration date.

The distinguishing efficiency of different food spoilage profiles on the principal component diagram can be further improved using sensors with different selectivities. This is inversely proportional to the correlation of the response of different sensors of the array with each other. Greater efficiency can be achieved if the sensors in the array provide a response of different polarities and amplitudes. For instance, this can be made by combining OFETs with p- and n-type organic semiconductors in an array[32] or by including the other types of gas sensors into such arrays, such as metal−oxide transistors or electrochemical cells.

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CONCLUSIONS

The electronic nose based on the D2-Und-BTBT-Hex organic semiconductor OFET sensor array covered with TiO−, Zn− and Cu−TPP metalloporphyrin receptors show sensitivity to the gases released by protein-containing food decomposition, known to contain amine- and sulfur-containing compounds. Due to the relatively low sensor baseline drift if compared to a useful spoilage-related signal and the sensor's high sensitivity, the gases released during pork, chicken, fish, or milk spoilage at room temperature can be detected as early as the 5th h of storage. The reference bacteriological counting measurements done on the chicken samples have shown that the device is capable of detecting spoilage byproducts at the 4×10^4 CFU g^{-1} bacteria count level, which is 2 orders of magnitude below the safe consumption threshold. Using the cross-selective OFET sensor array with metalloporphyrin receptors and principal component analysis, the device was able to distinguish chicken from pork and milk, which allows selective freshness detection of these products stored together. Appropriate pretraining by comparison with the training dataset collected for selected products and/or gases leads to distinguishing spoiled food and the composition of the escaping gases. If the findings are confirmed at lower temperatures, the device described could be integrated into butcher fridges or special raw meat storage boxes in household fridges, where meat is stored unpacked. The packing would significantly lower any compound concentration, while some previous works report on spoilage detection using e-nose with packed food.[28] Furthermore, such spoilage profiles matched with bacteriological measurements are collected for varied food types and temperatures to notify users of the approximated storage time and can be used to remind them to either cook or discard specific products. Additionally, we have to note that the presented e-nose is potentially much more affordable than the commercial ones since all of the sensors are produced on a single substrate by easily scalable self-assembly methods, consume very low power, and the data acquisition is designed on very simple elements, which altogether with the presented results ensure its competitiveness with conventional electronic noses in the future.

EXPERIMENTAL SECTION

Food Spoilage Experiments. The device consisted of an array of OFET-based sensors with different receptor layers, a substrate heater with a temperature sensor, an air temperature sensor, a sensor of relative air humidity, a sensor chamber, an analog signal generating and measuring device, and a microcontroller, which operates with all of the electronics described elsewhere.[25] The experiments with food were conducted at the room temperature of +25 °C in the laboratory hood and had a duration of 25 h unless otherwise stated. The fresh food was purchased from the same supplier (Myasnov) in the early morning before each experiment. The food samples of 50 g each were put into a 0.6 L container as single pieces or were poured on the bottom in the case of milk. During all of the experiments, a commercial Sensirion SHT-35 T/RH sensor was used inside the sensor chamber to compensate for the effect of environmental conditions on the OFET-based sensor readings.

Bacteria Counting Experiments. Two pieces (50 g each) of fresh chicken breast were placed into a sealed food container immediately after purchase in the early morning and stored at room temperature. Then, it was sampled by four pieces (5.0 g each) at 0, 5, 10, and 24 h storage time. The meat sample selected was finely chopped and poured into 1.0 mL of distilled deionized water. The resulting extract was diluted 10 and 10^4 times with distilled deionized water to get two samples each and then put onto Petri dishes with agar as a nutrient medium for bacterial growth. The diluted extract (100 μL) was rubbed into the agar and placed in a climate chamber at 40% humidity and 37 °C for 24 hours each. As a reference experiment, 100 μL of the same distilled deionized water was also rubbed into the agar and placed in a climate chamber at the same conditions. After that, the dishes were photographed, and colonies were counted using ImageJ software. All of the values were normalized to relative CFU per gram.

Data Analysis. To use machine learning with the data provided by the electronic nose (see the Supporting Information, Tables S1 and S2), it was further analyzed using the python script and sci-kit learn 1.0.2 library with the principal component, linear discriminant, and local linear embedding methods as well as logistic regression.[33] Prior to transformation, standard scaling was performed on the dataset.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsomega.2c06386.

Data on bacteria counting measurements, raw electronic nose data, and results of additional analysis methods (PDF)

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Author Contributions

S.A.P., E.V.A., and D.S.A. conceived the idea and designed the experiments. D.S.A., V.P.G., and D.S.K. performed the experiments. V.P.G. fabricated the sensor array. A.A.A. designed and fabricated the device and experimental setup. V.P.G. and D.S.K. conducted the bacteria counting experi-

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ments. D.S.A. performed data analysis and interpretation. E.V.A. and S.A.P. supervised the project. D.S.A., E.V.A., and S.A.P. wrote the paper. All of the authors contributed to the writing of the manuscript and have given approval to its final version.

Notes

The authors declare no competing financial interest.

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