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Research Article

Electronic Detection of Pesticide Residues on Cherry Fruits

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Abstract:

Agriculture is one of the most basic needs for the human generation to be sustained. In agriculture, pesticides are indispensable to increase productivity and prevent crop losses. Some of these drugs are pesticides that protect the plant from harmful insects, fungi and rodents, while others are herbicides that protect it from weeds. However, if these chemicals penetrate people as much as they ensure food safety, they can be harmful to various diseases in the long term. After these pesticides are applied to agricultural products, they decrease to safe levels after 3-10 days, depending on the type and dosage of the drug. Long-term exposure to these chemicals can cause several health problems, such as cancer, hormonal disorders, neurological diseases and immune system weakness. This study investigated whether the collected and pesticide-free cherry could be separated by smell after the pesticide was applied to it without passing the mentioned time. Cherries were collected from cherry trees that had never been sprayed; pesticide was sprayed on these trees, and cherries sprayed with pesticide were collected a day later. An electronic nose consisting of 11 very affordable gas sensors has been made for the study. The electronic nose took one hundred pieces of odours, including different amounts of cherries with and without pesticides. Various attributes of these data have been extracted. Among the four classification algorithms, the Extra Trees Classifier has the most successful results with 94.30% classification accuracy, 93.00% sensitivity and 95.60% specificity classification success. The ability to detect the pesticides on the fruit with an electronic device is important for monitoring human health through food inspections.

1. Introduction

It is known that in the history of humanity, people engaged in the first agricultural activities after moving from a hunter-nomadic life to a settled life. M.D., it is known that people started to domesticate animals such as goats, sheep, and cattle for the first time in the Neolithic Revolution around 10,000 years ago, and they also cultivated wheat, barley, and legumes [1]. Later, M.D. 4500-M.S., it has been determined that corn and rice were planted during the Ancient Agricultural process for over 500 years, and irrigation techniques were developed with the beginning of the use of ploughs [2]. After that, M.S. from the 500s to the 1800s, agricultural products diversified, trade in agricultural products increased, and agriculture was now consciously carried out more scientifically [3]. It covers the years from the 1800s to the 1980s, decently known as the modern agricultural or industrial revolution. In recent years, the use of agricultural machinery such as tractors and combine harvesters has started. Again, in these years, fertilizers and spraying began, and more modern irrigation techniques were used [4]. After 1980, it is now considered modern agriculture, where technology has penetrated agriculture. Genetic engineering has been developed in modern agriculture and has started to play with food genetics. Agricultural control and working methods have developed with the use of sensors. In addition, organic agriculture has been formed due to the threat posed to human health by food products grown using pharmaceuticals [5]. With the development of technology, spraying and irrigation have now started to be carried out by drones [6].

Electronic noses are used as electronic systems that can recognize odours that have been introduced to them before [7]. There are studies conducted on the quality [8], species [9], flavours [10], and spoilage [11] of a wide variety of foods with electronic noses. There are also studies on the use of electronic noses when growing agricultural

products. Health monitoring studies of plants [12], disease detection studies of plants [13] and quality control studies of products [14] have been carried out with electronic noses.
The studies conducted using e-nose related to cherry, which formed the basis of this study, are as follows: Cherry juice was fermented using various lactic acid bacteria, and the resulting aroma profiles were determined by an e-nose and gas chromatography-ion mobility spectrometry [15]. Again, in another study, aroma profiles of "Ferrovia" type cherries, packaged in cold storage or a high CO2 environment to maintain their freshness and quality, were determined using e-nose [16]. However, in another study, the degree of ripeness of cherries was determined by their smell [17]. In addition to these studies, in another study that is the same as our study topic, cherries with pesticide applied and cherries without pesticides were distinguished from each other by using an electronic nose. In this study, 48 cherries, 12 at four different maturity stages, were collected from cherry trees treated with the agricultural pesticide Diazinon mixed with water at a rate of 1/1000, and the same amount of cherries was collected from cherry trees that were not sprayed with pesticides. They were immediately sent to the laboratory for analysis in an insulated box containing ice. Here; their odours were captured by an electronic nose consisting of MQ3, MQ5, MQ9, MQ135, TGS2620, TGS2610, TGS2611, TGS813, TGS822 and TGS2602 gas sensors. Here, 96 cherries were individually scented. Each scent cycle lasted 750 seconds. After these data were divided into training-test sets as 75-25%, they were separated from each other with 72-86%, 93.75% and 100% accuracies, respectively, using Principal Component Analysis (PCA), Linear Discriminant Analysis (LDA) and Artificial Neural Network (ANN) methods. [18]. In this study, cherries with and without pesticides were separated from each other with full accuracy using an electronic nose, but in this study, cherries with pesticides were obtained as soon as the pesticide was sprayed on the cherries. When smelling pesticide-laced cherries, no damping period of the pesticide was expected, and therefore, it can be interpreted as smelling the pesticide directly in this study. Additionally, the sniffing cycle time with the electronic nose lasted 750 seconds.
The environmental and economic costs of pesticides have been examined, and their negative effects on human health, ecosystems, and the agricultural economy have been revealed in a study [19]. In parallel with this, in another study, although pesticides are used due to their effects on increasing productivity, their harms to human health and the environment were discussed [20]. Similarly, in another study on the subject, the harm of pesticides to the ecosystem, the threats their residues pose to food safety, and their harm to human health were mentioned [21]. As a result of a study conducted in 2013, it was determined that pesticide exposure increased the risk of cancer and cancer rates increased in people exposed to pesticides [22].
Pesticides are indispensable drugs today for the efficiency of agriculture. However, a "pre-harvest interval" period must be waited after pesticide application. This period varies depending on the type of pesticide applied and the dosage applied. It has been determined that this period is generally 3-10. Only after this period will the product be sprayed with pesticides become safe for human consumption. For this reason, fruits and vegetables that have been sprayed with pesticides should not be offered to people for consumption without waiting for this necessary period [23].
The process of worming cherries begins when the cherry fly (Rhagoletis cerasi) lays its eggs inside the fruit in early or mid-June when the cherries begin to ripen. The larvae that hatch from the eggs feed inside these cherries, causing the cherries to become wormy. Spraying should be done before these flies lay their eggs [24]. Therefore, farmers spray their cherries when they begin to ripen, but customers do not know when they take them to the market. When people shop, they want to make sure that the fruits and vegetables they buy are as freshly picked as possible. However, they cannot know whether the product they purchase has a pesticide effect. Institutions and organizations that carry out food inspections must be able to detect products put on the market without waiting for the above-mentioned period.
This study aimed to detect cherries sprayed with pesticides the day before and cherries without pesticides using an electronic nose in 60 seconds without any processing. As a result of the study, a device that detects pesticides on fruits and vegetables, especially cherries, can be performed. The output of the study may be useful for the inspection of fruits and vegetables sold as commercial products.

2. Material and Methods

2.1. Sample preparation

Cherries sprayed with pesticide and cherries without pesticide were collected from an orchard in Çorum. The fruits were collected without any physical damage. The cherries were picked in early June without any worms or pesticides. Then, pesticides were sprayed on these cherry trees to protect them from the cherry fly, which causes worms to lay eggs on cherries. This pesticide was prepared by mixing and dissolving 20 g of the drug called Korban 25W into 10 litres of water, then sprayed on cherry trees. The cherries sprayed with pesticide were picked a day later.

2.2. Electronic Nose System

For this study, a sensor block was made with 11 gas sensors that can be purchased at very affordable prices in the market. Gas sensors were used with their own kits. The gas sensors used are: MQ-2, MQ-3, MQ-4, MQ-5, MQ-6, MQ-7, MQ-8, MQ-9, MQ-131, MQ-135, MQ-137.

The sensor block was placed in a box with a lid, into which the samples could be placed, and the sensor cables were taken out of the box through an airtight hole. The electrical data taken from the gas sensor kits were converted into digital data via two Arduino Uno cards and transferred to the computer. The software running this recording system has been prepared with the LabVIEW (2016) program. The e-nose system made for the study is shown in Figure 1.

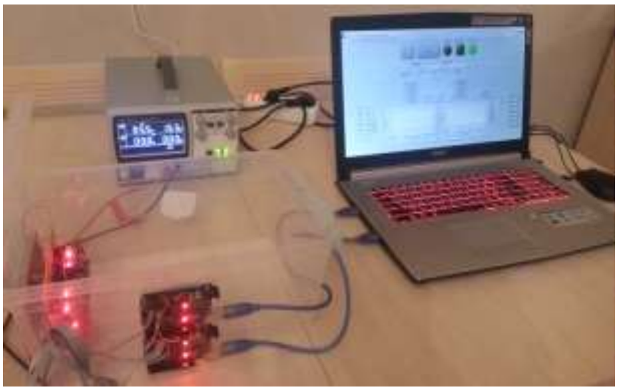

***Figure 1.** Prepared e-nose system*

2.3. Data Collection and Classification

The cherries obtained were in two groups: cherries with pesticides and without them. Within 3-8 hours after these cherries were collected, ten odours were recorded from 100-200-300-400-500 grams each, with the e-nose. Figure 2 shows an image of 200 g of cherries without pesticides.

***Figure 2.** 200 grams of cherries without pesticides*

All measurements were made between 12.00 and 17.00, under 23-26 °C temperature and 50-70% humidity conditions. Here, a total of 50 cherries without pesticide odour data and 50 cherries with pesticide odour data were recorded on the computer. The number of samples from which odour records were taken is given in Table 1.

***Table 1.** Recorded odour samples*

	Cherry without pesticide	Cherry with pesticide
100 g cherry	10	10
200 g cherry	10	10
300 g cherry	10	10
400 g cherry	10	10
500 g cherry	10	10
Total	50	50

The sniffing cycle begins by placing the cherry samples in a previously ventilated e-nose odour box, then closing the lid of the box and starting the LabView software. E-nose odour recording time was 60 seconds, and data was taken from the sensors ten times per second. 11x601 pieces of data were obtained in each odour recording. As clearly stated in Table 1, 100 separate cherry scent records were taken, and a three-dimensional data matrix with a total size of 100x11x601 was obtained.

The statistical attributes whose formulas are given below (1-4) of these data have been extracted.

$$x_{mean} = \sum_{x=1}^{s} \frac{x_1 + x_2 + \cdots + x_s}{s} \tag{1}$$

$$x_{std} = \sqrt{\frac{(x_1 - x_{mean})^2 + \cdots + (x_s - x_{mean})^2}{s-1}} \tag{2}$$

$$x_{sum} = x_1 + x_2 + \ldots + x_s \tag{3}$$

$$x_{median} = \begin{cases} x_{\frac{s+1}{2}} & \text{if s is odd} \\ \frac{1}{2}\left(x_{\frac{s}{2}} + x_{\frac{s}{2}+1}\right) & \text{if s is even} \end{cases} \tag{4}$$

Here, x_{mean} is the mean value of a trial, x_{std} is the standard deviation value of a trial, x_{sum} is the total value of a trial, x_s is the last value of a trial, s is the value number of a trial, x_{median} is the value in the middle of a trial.

In the feature extraction process, four different feature extraction processes were applied to the data of 11 gas sensors and a total of 44 features were obtained. In the study, the most frequently used classification algorithms today, which are generally tree-structured, are used, such as Random Forest Classifier, Extra Trees Classifier, Decision Tree Classifier and k-nearest Neighbor (kNN) Classifier. As it is frequently mentioned in the literature, since tree classifiers decide which feature and how many features to use by using random subsets of all features, in this study, 44 features were made available to classifiers without feature selection [25].

After feature extraction, the data was divided into 60% training, 20% validation and 20% test data. In the classification process, classifiers are trained with training data, adjustments are made with validation data, and then these classifiers are tested with test data. The result gives the performance of the classification. The following classification criteria (5-7) were used to evaluate the performance of the classifiers [26]:

$$CA = \frac{CCT}{TT} x100 \tag{5}$$

$$SE = \frac{TP}{TP + FN} x100 \tag{6}$$

$$SF = \frac{TN}{TN + FP} x100 \tag{7}$$

CA: Classification Accuracy, SE: Sensitivity, SF: Specifity, CCT: Correctly Classified Trials, TT: Total Trials, TP: True Positive, TN: True Negative, FP: False Positive, FN: False Negative.

For the reliability of the classification study, the above-mentioned classification process was carried out 100 times for different data sets by randomly selecting the training-validation-test sets. The arithmetic average of these 100 classification results was accepted as the classification success. The working principle of classification algorithms is also shown in Figure 3.

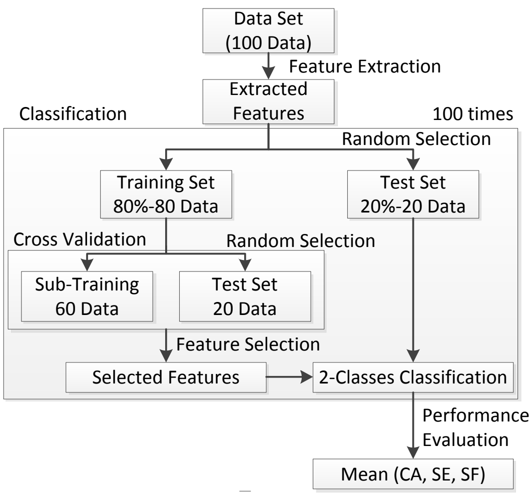

Figure 3. *Classification Flow Diagram*

The kNN classification method is a widely used and highly effective technique in classification studies. In kNN classification, distances between data points are calculated. Test data is classified based on the majority of its k nearest neighbours. The optimal value of k is determined using the cross-validation method [27].

The decision tree classification method is a tree-like decision model; it creates decision rules by separating the data into branches and classifying them. Each internal node of the tree represents a feature, each branch represents a decision rule, and each leaf node represents an outcome or class label. The tree is created by iteratively splitting the dataset according to the feature that provides the best split, using criteria such as the Gini coefficient or entropy. This process continues until the tree classifies the training data perfectly or reaches a predefined stopping condition. Decision Trees are easy to interpret and visualize but can be prone to overfitting, especially when they are deep and complex [28].

Random Forest Classification is a widely used classification method that makes decisions using multiple decision trees. Each tree is trained on a random subset of data with randomly selected features. A majority vote of the trees makes the final classification decision. This approach reduces the risk of overfitting due to the diversity of trees and increases generalization ability [29].

The Extra Trees classification method is similar to the Random Forest method except that it trains every tree in the entire dataset. In this method, split points are chosen completely randomly. As a result, trees are more diverse, and training times are generally faster. This method also prevents overfitting due to the high level of randomness. While splitting criteria can be based on the Gini coefficient or entropy, actual splitting points are chosen randomly [30].

3. Results and Discussions

In this study, cherries with pesticides and cherries without pesticides were distinguished from each other by using an e-nose from only their odour. An e-nose using 11 gas sensors was made for the study. The smell of 100 grams (g), 200 g, 300 g, 400 g and 500 g cherries with and without pesticides was recorded ten times per each with the e-nose, so a total of 100 samples of cherry odour data were obtained. A value matrix of 601x11x100 size was created by recording data from 11 different gas sensors for 60 seconds.

Four different features of the data obtained from 11 gas sensors were extracted, and the 44 features obtained were classified with four classification algorithms. At the end of 100 different classifications made with randomly selected training, verification and test sets, the average values of the obtained performance parameters, such as classification accuracy, sensitivity and specificity, are given in Table 2.

Table 2. *Classification Results According to Classifiers*

Classification Algorithm	CA (%)	SE (%)	SF (%)
Extra Trees	94.30	93.00	95.60
3-kNN	90.15	86.10	94.20
Random Forest	89.90	89.10	90.70
Decision Tree	84.95	83.40	86.50

The importance of all features used in classification according to the Random Forest Classifier is given as percentages in Figure 4. The number k is determined as 3 using the cross-validation method in the Nearest Neighbour classification algorithm. The average confusion matrix of 100 classifications for the Extra Trees classification algorithm test data, which gives the highest classification accuracy in classifications, is given as a percentage in Table 3.

Table 3. *The Confusion Matrix of the Extra Trees Classification (%)*

Classification Accuracy: 94.30%		**Predicted**	
		Cherry with pesticide	**Cherry without pesticide**
Real	**Cherry with pesticide**	96	4
	Cherry without pesticide	7	93

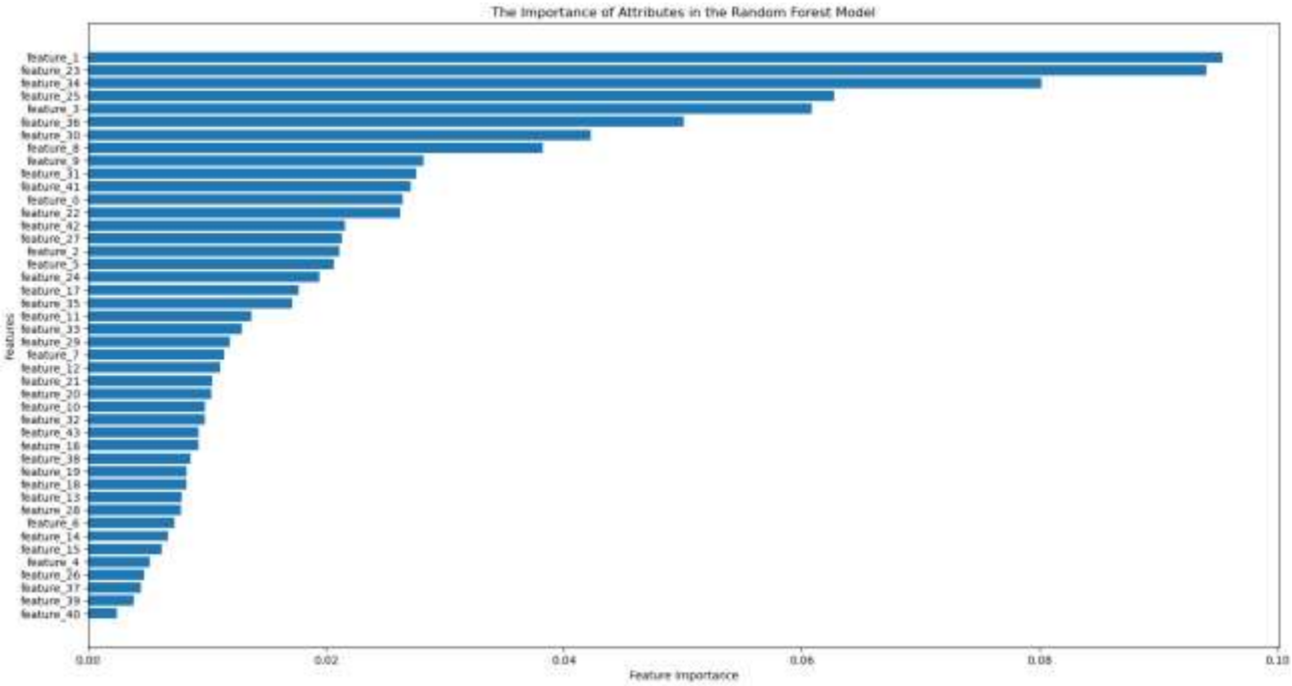

Figure 4. *The importance of features*

Table 4. *The Importance of Features and Gas Sensors*

	MQ2	MQ3	MQ4	MQ5	MQ6	MQ7	MQ8	MQ9	MQ131	MQ135	MQ137
Mean value	2.64	9.54	2.11	6.09	0.52	2.07	0.73	1.14	3.82	2.82	0.99
Standard deviation value	1.37	1.11	0.78	0.67	0.62	0.93	1.77	0.83	0.83	1.03	1.04
Sum value	2.62	9.40	1.95	6.28	0.47	2.13	0.78	1.19	4.22	2.76	0.99
Median value	1.29	8.02	1.72	5.01	0.44	0.86	0.38	0.24	2.71	2.16	0.93
Total sensor effect %	7.93	28.07	6.56	18.05	2.05	5.99	3.66	3.40	11.59	8.78	3.94

Which gas sensor data the features shown in Figure 4 belong to is given in Table 4, along with their usage percentages. Here, the effect of gas sensors on the result is clearly seen. Considering the usage percentages, it can be said that the important sensors for this study are MQ-3, MQ-5, MQ-131, MQ135 and MQ-2, respectively.

The low number of data usage is one of the negative aspects of the study. In addition, the use of a single type of pesticide can also be considered as another negative aspect of the study. In future studies, studies with different pesticides and different fruits and vegetables will be useful in this area.

4. Conclusion

In this study, an e-nose with eleven gas sensors was made. The odours of cherry samples with and without pesticide, in different amounts, were taken with an e-nose. Four different features of a total of a hundred odour data were calculated, and these were classified a hundred times using different training-test data selections by four different classification algorithms. Test data was best classified using the Extra Trees Classification Algorithm with 94.30% CA, 93.00% SE and 95.60% SF classification performance. While the MQ3 gas sensor provides the most valuable data, the least important data is produced by the MQ-6 gas sensor. As a result of the study, pesticides on cherries were detected with high accuracy through e-nose with the proposed method.

Author Statements:

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- **Ethical approval:** This research is not related to either human or animal use.
- **Conflict of interest:** The author declares no conflicts of interest.
- **Data availability statement:** Since the author is conducting other studies with this data, the data has not yet been shared with the public.

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