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A Comparison of the Multivariate Calibration Methods with Feature Selection for Gas Sensors' Long-Term Drift Effect

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Abstract: In many electronic nose applications where gas sensors utilizing for a long time, there is an undesirable drift effect on the sensors, which affects the classification quality negatively. Although the sensor drift is inevitable, it is possible to reduce this effect with the calibration transfer methods. This paper presents a comparison study of various multivariate standardization methods to facilitate an effective calibration way on a comprehensive dataset, which is reachable on-line. In this study, three methods applied: direct standardization (DS) orthogonal signal correction (OSC) and piecewise direct standardization (PDS). In addition, these three methods are applied data, which consisted of selected features. The results have shown that the classification success has increased with multivariate calibration technique applied to the selected features. The results also demonstrate that using the best features in the signal processing part can play an important role for the calibration success. This outcome may lead to a new perspective for the future works.

Gaz Sensörlerinde Uzun Süreli Sapma Etkileri için Öznitellik Seçimi ile Çok Değişkenli Kalibrasyon Yöntemlerinin Karşılaştırılması

Anahtar Kelimeler
Kalibrasyon transferi
Öznitelik seçimi
Gaz sensörleri
Çok değişkenli sapma düzeltme
Standartlaştırma yöntemleri

Özet: Gaz sensörlerinin uzun süreli kullandığı elektronik burun uygulamalarında, sensörler üzerinde istenmeyen bir sapma etkisi meydana gelir. Bu etki verilerin analizinde çok etkin olup ölçüm sonuçlarının doğru analizini engellemektedir. Bu durum kaçınılmaz olsa da, istenmeyen etkiyi kalibrasyon transfer yöntemleriyle azaltmak mümkündür. Bu çalışma kapsamında, çevrimiçi olarak erişilebilen bir veri seti üzerinde çok değişkenli kalibrasyon yöntemlerinden olan doğrudan standartlaştırma (direct standardization-DS), ortogonal sinyal düzeltimi (orthogonal signal correction-OSC) ile parçalı doğrudan standartlaştırma (piecewise direct standardization-PDS) yöntemleri uygulanmıştır. Ayrıca yine bu yöntemler; aynı veri setinin öznitelik seçimi yapılmış haline uygulanmış ve sonuçların tümü karşılaştırılarak en başarılı yöntem, sınıflandırma başarı oranlarına bakılarak bulunmaya çalışılmıştır. Sonuçlara göre; öznitelik seçimi yapılan veri setine uygulanan kalibrasyon yöntemleri, ham veri setine oranla daha başarılı olmuştur. Böylece daha az veri ile kalibrasyonun başarılı bir şekilde gerçekleşebileceği gözlemlenmiştir. Bu çıktının gelecek çalışmalarda yeni bir perspektif yaratacağı düşünülmektedir.

1. Introduction

In the last few decades, there has been significant improvement of electronic nose (e-nose) devices, which are consisting of gas sensor arrays. Whilst gas sensors are operating for a long time, the sensors' responses change in time without any control, and it is called sensor drift. There are two types of sensor drift: short-term drift (second order drift) and long-term drift (first order or real drift). The short-term drift occurs because of vicinity factors. For instance, temperature, humidity, thermal and memory effects. These factors can keep under control. On the other hand, long-term drift cannot. The long-term drift is inevitable, because it occurs at sensors' internal structures. As known, the chemical processes are not reversible. For robust measurements, sensors' responses need to stabilize over time. Hence, the calibration methods are proposed to cope with the drift effect (Pearce, 2002).

Different ways have been tried for getting rid of sensor drift with a calibration model in the literature so far. To correct the drift effects, some of studies are utilized the univariate calibration techniques, some of them are utilized the multivariate calibration techniques. For the univariate technique, every variables of the master instrument matches the variables of the slave instrument for fitting a curve. After calculating two variables of the curve slope and intercept point, the univariate calibration can be used to get results. However this technique seems easy for usage, it does not appropriate solution considering gas sensors' applications. So in this study, the multivariate calibration techniques are focused.

So many studies with different ways and proposed methods had been done in the literature before. However, a reviewed article is one of the most comprehensive one (Feudale, 2002). Several methods were reviewed in this article, and some of the methods were studied with near infrared (NIR) data. Direct standardization (DS), piece-wise direct standardization (PDS), artificial neural network (ANN), maximum likelihood principal component analysis (MLPCA), and positive matrix factorization (PMF) methods were investigated. According to the work, PDS might be the best solution for complex systems. Moreover, ANN could be easily applied, but it suffered from overfitting problem. As a result of this article, for choosing the best technique there is no perfect guideline.

Another study about calibration transfer methods is Pereira and his team's study. In this work, in order to specify some ingredients in gasoline three NIR spectrometers were studied (Pereira, 2008). DS, PDS, orthogonal signal correction (OSC), reverse standardization (RS), piecewise reverse standardization (PRS), slope and bias correction (SBC), and model updating (MU) methods were compared to each other and researchers claimed that RS was the best method. Furthermore, MU was the best way, when the transfer samples could not storage practically.

Although some methods are using for interpreting the signals, e.g., principal component analysis (PCA) and partial least squares (PLS). These methods were also using for the component correction (Tomic, 2002). It is because; component correction is based on OSC. With using 39 gas sensors for detecting four gases, and after logging the recordings for two months, multiplicative drift correction (MDC), PCA and PLS based component correction methods were compared, and the best root mean square error prediction (RMSEP) value could get by PLS based component correction method (Artursson, 2000).

Galvãoa and his team proposed a new method for the calibration transfer. The method differs from the related works because of the robustness. The authors presented a model which was consisting of a univariate procedure first and then a robust regression technique for building a multivariate calibration model. According to the results, the robust regression procedure was better than the univariate correction alone (Galvãoa, 2015).

Panchuk et. al. presented a method for e-nose applications which was based on the DS algorithm. In the paper, the calibration models between different analytical methods were proposed. As a results of the obtained multivariate regression model, samples were successfully transferred to the slave's instrument (Panchuk, 2017).

One of the most commonly used methods in calibration transfer area is PDS. Zhang et. al. introduced a new calibration process which is called sampling error profile analysis (SEPA) for optimization some parameters in the PDS algorithm. In PDS algorithm, in order to transfer the samples one instrument to another PLS with mean-centering was used. The proposed algorithm got better results than two other calibration transfer methods: SBC and spectral space transformation (SST) (Zhang, 2017).

According to the literature, the most successful and the popular methods seem DS, PDS and OSC (Artursson, 2000; Fernandez, 2016; Feudale, 2002; Galvãoa, 2015; Malli, 2017; Panchuk, 2017; Pereira, 2008; Tomic, 2002; Zhang, 2017). Just as the former literature's works, this study is also followed the trend, and compared their classification successes for different features. Because of the diversity of gas sensors and their applications, none of the studies in the literature could not give a guarantee solution for calibration. Nevertheless, they recommend possible solutions for specific applications. However, we beilive that our proposed method could implement any application where gas sensors are using.

2. Material and Method

2.1. Dataset

A comprehensive dataset, which is freely reachable on-line is used for this study (Vergara, 2012). The data was collected from sixteen metal-oxide gas sensors' for three years, and the dataset has 13,910 recordings total. The aim of the Vergara's study was to distinguish the six different components: ammonia, acetaldehyde, acetone, ethylene, ethanol, and toluene regardless of their concentration. In that study, eight features were extracted: the steady-state response of the sensors (the differentiation maximum value and minimum value of the sensor responses), normalized version of steady-state response, exponential moving average of both rising, and decaying portion of the sensor response for three different smoothing parameters.

The data collected for 36 months is not distributed uniform month by month. That is why, 10 batches are composed instead of categorizing 36 measurement sets. In this study, batch 7 and batch 10 are used. It is because that month 21, which means batch 7, has 3613 total recordings. On the other side, month 36, batch 10, has 3600 total recordings with every kind of gas. The intention of this study is to build a calibration model via these samples. The goal is to train the model on batch 7 and to test it on batch 10. Because, the time interval between two batches is 15 months and it is enough time to investigate drift effect on sensors' surface. The assumption is that the data of batch 7 is obtained from master instrument, and the data of batch 10 is obtained from slave instrument.

2.2. Feature selection

Feature selection has an important role for the machine learning systems. The main aim of the feature selection is that neglect some input features that effects output minor. In other words, the feature selection is applied to reduce the dimension of the data. It also means that data become more understandable. Furthermore, the irrelevant input features cause greater computational cost. Namely, the feature selection is essential.

There are many ways to select the features from the data. In this work, the sequential selection algorithm was used. It starts with computing all feature subsets, which consist of only one input feature. So, it measures the leave-one-out cross validation error of the one-component subsets (Chandrashekar,2014).

2.3. Methods

The need of the calibration model was discussed earlier. The general idea of all the calibration methods is that modelling variations between two instruments and diminish it. Creating a calibration model consisting of four fundamental steps.

1. Selecting the transfer samples from both master and slave instruments. D optimal, E optimal, and Kennard-Stone (KS) algorithm can be performed to choose the transfer samples.
2. Generating a mathematical model using the transfer samples.
3. Transform the samples of the slave instrument's to the responses of the master instrument's.
4. Eventually, the prediction model is trained on master instrument's transfer samples, and it is ready for mathematical manipulating in order to reduce drift effect.

Among the different calibration models, the above model is selected and DS, PDS and OSC methods are used for transforming the slave instrument's samples to the master instrument's response. These methods are applied to the slave instrument's data for the correction. An illustration of this correction can be seen in the Figure 1 (Haugen, 2000).

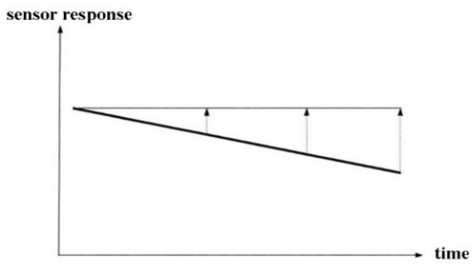

Figure 1. Drift affect on a sensor

Direct standardization (DS) is one of the multivariate calibration techniques, proposed by Wang et al. This method uses a transformation matrix, T, provides a linear relationship between two instruments' responses. As Y_1 is a master instrument's standardized samples matrix and Y_2 is the slave samples matrix, which is consisting of samples that was going to manipulate in order to adjust drift effect. T transformation matrix can be calculated as Equation (1).

$$\mathrm{T} = Y_2^{+} Y_1 \tag{1}$$

where Y_2^{+} is a pseudo-inverse of Y_2. After calculating T, instrument responses can be predicted by multiplying new samples transpose matrix with T. As seen, this method presents a linear relationship. Therefore, it is not an excellent technique. Besides, it has over-fitting problem, unless there is enough transfer samples (Wang, 1991).

Piecewise direct standardization (PDS) is an also the multivariate calibration technique (Wang, 1991). This method is similar to DS except it uses windowing technique. Every transfer samples of the master instrument correspond to slave instrument's transfer samples, which are located in a sliding window.

For example, the number of the sensors be n, *i* be the any sensor, and r be the response of the standardization samples measured on the master instruments is corresponding to the sensors located in the window around *i*th sensor. For getting a sub-transformation matrix of ith sensor t*i*, Equation (2) is applied where R is the response matrix of the transfer samples.

$$\mathrm{r}i = \mathrm{R}i \ \ \mathrm{t}i \tag{2}$$

Afterwards, for all the sensors, sub-transformation matrices compiled in one big matrix, which is illustrated as symbol T.

$$\mathrm{T} = \mathrm{diag}\,(t_1^T, t_2^T, t_3^T, \dots, t_n^T) \tag{3}$$

With calculated matrix T, the calibration model can be applied in order to remove differences between two devices.

OSC is another method, and it generally uses for pre-processing (Wold, 1998). The reason is that its objective is to remove the noise by finding the components, which are orthogonal to the data. Nowadays, OSC is also using for the multivariate calibration and the idea is the same: remove vectors, which are orthogonal to both instruments to make the model more transferable (Feudale, 2002).

The algorithm used in OSC is very much alike NIPALS algorithm, which used in PCA and PLS. In this algorithm, the weight vector (w) is updating, in accordance with the score vector (t). The score vector can be calculated as Equation (4), and the standardized data's expression of X is given as Equation (5).

$$t = X.w \tag{4}$$

$$\mathrm{Xosc} = \mathrm{X} - \sum_{i=1}^{n} Ti.Pi' \tag{5}$$

where T is the classes of gases, P is the loadings matrices, and n is the OSC factors, which means that how many time this procedure is applied.

In this work, a comparison of some calibration methods are made and a different way for standardization process is tried. The assumption is that if the best features selected, the classification success may increase regardless of which calibration model is applied or unapplied. The attemption is to select best features, which means that the highest classification success with chosen features. DS, PDS and OSC are applied to the comprehensive dataset. Besides, these three different methods, and the raw data with the best features are tried. As mentioned above, the sequential selection algorithm is used in the feature selection process. The classification success is evaluated by k-NN (k-nearest neighbor) algorithm, and k parameter is determined with the cross validation method.

To achieve the goal, two batches are considered: batch 7 and batch 10. For the analogy, the batch 7 is the master and the batch 10 is the slave instrument. Therefore, two groups are analyzed. Group 1 is consisting of chosen training and chosen test data from batch 10. Group 2 is consisting of chosen training data from batch 7 and chosen testing data from batch 10.

3. Results

The attemption is to increase the classification success began with finding the success of raw data, the direct standardized data, the standardized data by orthogonal signal correction, and the piece-wise direct standardized data. Afterwards, the finest features are determined and same methods are applied again, but this time along with the selected features. The classification successes of the raw data, the direct standardized data, the standardized data by orthogonal signal correction and the piece-wise direct standardized data are given in Table 1. As shown in Table 1, the most efficient calibration method is PDS for both groups. On the other hand, the classification successes of same adjustment with selected best features of data are given in Table 2. As shown in Table 2, the most effective calibration method is again PDS for two groups.

According to outcomes of the tables, it can be said that selecting features has a great impact on datasets, especially for group 2. The performance is improved for all adjustments except PDS. However, the success of PDS's and PDS with selected features' percentages are 95,41 and 94,81 in a row, and the difference between them seems minor.

Table 1. Classification Success for Calibration Methods

Dataset		Success (%)			
Train Data Set	**Test Data Set**	**Success 1[a]**	**Success 2[b]**	**Success 3[c]**	**Success 4[d]**
Batch10	Batch10	87,68	60,83	95,41	87,73
Batch7	Batch10	64,12	63,33	67,91	64,12

[a.] success of raw data, [b.] success of DS, [c.] success of PDS, [d.] success of OSC

Table 2. Classification Success for Calibration Methods with Selected Features

Dataset		Success (%)			
Train Data Set	**Test Data Set**	**Success 4[e]**	**Success 5[f]**	**Success 6[g]**	**Success 7[h]**
Batch10	Batch10	88,14	88,70	94,81	88,14
Batch7	Batch10	72,87	77,17	75,69	72,87

[e.] success of raw data with selected features, [f.] success of DS with selected features, [g.] success of PDS with selected features, [h.] success of OSC with selected features

For representing classification successes more clear, confusion matrices for raw data, and PDS with selected features technique for all six gases are demonstrated in Table 3 and Table 4 respectively. By taking these results, Table 5 and Table 6 are created, and they illustrate the sensitivity vs. specificity of the six gases. The most drifted data according to Table 3 and 5, belongs to ammonia and toluene gas. It can be seen clearly from Table 4 and 6, selecting features increases the sensitivity of the data.

Table 3. A Confusion Matrix for Raw Data

		Predicted Values					
		A[i]	***B[j]***	***C[k]***	***D[l]***	***E[m]***	***F[n]***
Actual Values	***A[i]***	358	0	0	22	0	0
	B[j]	0	331	0	0	1	0
	C[k]	0	51	118	0	0	0
	D[l]	51	40	0	444	0	3
	E[m]	0	0	2	0	324	2
	F[n]	0	0	0	38	65	323

Table 4. A Confusion Matrix for PDS with Selected Features

		Predicted Values					
		A[i]	***B[j]***	***C[k]***	***D[l]***	***E[m]***	***F[n]***
Actual Values	***A[i]***	296	0	0	22	0	0
	B[j]	0	338	2	0	0	0
	C[k]	0	21	357	0	0	0
	D[l]	64	0	0	337	0	0
	E[m]	0	0	0	0	360	0
	F[n]	0	1	1	1	0	360

[i.] Ethanol, [j.] Ethylene, [k.] Ammonia, [l.] Acetaldehyde, [m.] Acetone, [n.] Toluene

Table 5. Sensitivity and Specificity for Raw Data

Gas	**Specificity (%)**	**Sensitivity (%)**
Ethanol	97,16	94,21
Ethylene	95,06	99,70
Ammonia	99,90	69,82
Acetaldehyde	96,33	82,53
Acetone	96,42	98,78
Toluene	99,71	75,82

Table 6. Sensitivity and Specificity for PDS with Selected Features

Gas	**Specificity (%)**	**Sensitivity (%)**
Ethanol	96,52	93,08
Ethylene	98,79	99,41
Ammonia	99,83	94,44
Acetaldehyde	98,69	84,03
Acetone	100	100
Toluene	100	99,17

4. Discussion and Conclusion

Although gas sensors' drift is unavoidable, it has multiple solutions to eliminate this problem. The calibration transfer methods are useful to get rid of this drift effect. In this context, a significant improvement is achieved by using the raw data, DS, OSC and PDS with the sequential selection algorithm. The results of the study have pointed out that selecting the best features also can be a technique for calibrating alone. The important point here is to select the features, which are least effected from the drift. This may be the motivation for the future works. On the other side, when it is combined with DS, OSC and PDS methods, the best classification success value can get with PDS. So far, lots of calibration transfer techniques are proposed by many researchers but to the best of our knowledge, creating a model which is consisted of selected features for calibration is a new perspective. So, the contribution of this study is to be a guideline for future studies as well as to achieve more success in less time. Once and for all, the comparison in this study with or without feature selection has a potential to use in many machine learning applications.

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