AIR EMISSION MONITORING FROM WASTE INCINERATION/CO-INCINERATION INSTALLATIONS USING SURROGATE PARAMETERS

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Abstract: *Industrial emission directive (Directive) gives the obligation of continuous monitoring of some emissions into air for the incineration/co-incineration of waste by direct measurement. Conclusions for best available techniques for waste incineration, equally applied on co-incineration, allow the use of surrogate parameters instead or in combination, if this proves to be equivalent or better scientific quality than direct emission measurement. Surrogate parameters are measurable or calculable quantities closely related, directly or indirectly, to conventional direct measurements of emissions. The accent is on models that by combining more surrogate parameters could model emissions as well as valuation of the results of such models according to the requirement of Directive. The application considered aims environmental permitting.*
Keywords: *continuous monitoring, surrogate parameters, validated values*

Received: 19.12.2023. / Accepted: 18.01.2024.
Published online: 31.01.2024.

Professional paper
DOI:10.37023/ee.10.1-2.4

1. INTRODUCTION

Air monitoring emission from waste incineration/co-incineration installations is treated by Industrial emission directive (2010/75/EU) (Directive), Annex VI, Part 6, Point 1.3. **Equation 1** for direct measurement monitoring (validated values) is:

$$V_{1/2\,hr} = E_{mj} - t_{95,df} \cdot s \cdot \sqrt{\tfrac{1}{n}} \quad (1)$$

with confidence interval restriction according to limit emission values (GVE) given for daily average:

$$t_{95,df} \cdot s_{im} \cdot \sqrt{\frac{1}{n}} \leq p \cdot GVE \; (2)$$

In **Equation 2** for estimation of population standard deviation of measurement results one uses symbol s_{im}, and symbol s will be further used for estimation of population standard deviation of surrogate parameters model results.

According to Directive, the sample size for measurement is allowed to be *n=1*, and **Equation 2** becomes **Equation 3**:

$$t_{95,df} \cdot s_{im} \leq p \cdot GVE \; (3)$$

where p are coefficient in table from the Directive, Annex VI, Part 6, Point 1.3, $V_{1/2\,hr}$ validated values from direct measurement or for model results (30 min average), t_{95}, $t_{95,df}$ Student (*t)* distribution for 95% confidence level, E_{mj}, average value from direct measurement, df degree of freedom, *GVE* limit emission values, n_{im} sample size for direct measurement, *n* sample size for model, s_{im} estimation of population standard deviation from direct measurement, s estimation of population standard deviation from models.

According to the norm EN 14181 (Emissions from stationary systems– assurance of quality of automatic measurement systems (EN 14 181: 2014), it is allowed to use uncertainties in validation of direct measurements for automatic measurement systems:

$$V = E_{mj} - \left(\mu \cdot E_{mj}\right) \quad (4)$$

with limit for confidence interval:

$$\mu \cdot E_{mj} \leq p \cdot GVE \quad (5)$$

where are μ relative measurement uncertainty given as extended with 95% confidence level, E_{mj} average value for direct measurement (30 min average). Those equations for direct measurement monitoring also applies in surrogate parameters monitoring.

2. MONITORING EMISSIONS BY SURROGATE PARAMETERS

2.1. Basic frame for surrogate parameters monitoring

By Directive (Directive, 2010/75/EU) and conclusions on best available techniques (BAT), BAT Conclusions for large combustion plant (EU 2017/1442)), BAT Conclusions for the manufacture of glass (2012/134/EU) and BAT Conclusions for waste incineration (EU 2019/2010), the basic frame for surrogate parameters monitoring is given and always in some relation to direct measurement of emission. The relation between direct measurement and surrogate parameters is not always easy to understand. By article 15 of directive, it is allowed to use surrogate parameters for confirmation results of direct measurement. In the frame of that article surrogate parameters have subordinate role for the sake of confirmation of different monitoring frequencies then those given by BAT.

However, by Decisions on BAT conclusions on large combustion plant, BAT conclusions for the manufacture of glass and BAT conclusions for waste incineration, the role of surrogates was finally equalized with direct measurement of emissions into air. BAT conclusions for the manufacture of glass, technique BAT 7, allows to use surrogates together with direct periodic measurement for NOx, dust, and SOx instead of direct continuous measurement of those emission parameters, as also BAT conclusions for large combustion plants allows it without associated direct measurement for some types of plants (technique BAT 4) and instead continuous direct measurement. In BAT conclusions for waste incineration, techniques BAT 4 and BAT 5, it is allowed to use it without direct measurement of emission for regular and even for nonregular work.

The use of surrogate parameters according to article 15 of Directive is under further research because of always existing importance of confirmation for results of direct measurement results monitoring.

2.2. Monitoring emissions by predictive surrogate parameters

Predictive surrogate parameters are process parameters applied by models calculating emission values which are monitored, with important requirements on parameters to be also constantly measured (Brinkmann et al. 2018; Rumenjak 2021). Equation for validated model values with one sided confidence interval is:

$$V_{1/2\,hr} = \hat{y}_{1/2\,hr} - t_{95,df} \cdot s \cdot \sqrt{\frac{n+1}{n}} \ \ldots (6)$$

Symbol $\hat{y}_{1/2\,hr}$ is used for all predictive surrogate parameter models where $\hat{y}_{1/2\,hr}$ is predictive model for concentration (30 min average). **Equation 6** in this case is written without part for random error of input values what could be justified by narrower confidence interval on the lower side of the model.

Equation for the confidence interval limit for models must consider confidence interval for direct measurement as:

$$t_{95,df_{im}} \cdot s_{im}\sqrt{\frac{1}{n_{im}}} + t_{95,df} \cdot s \cdot \sqrt{\frac{n+1}{n}} \leq p \cdot GVE \ (7)$$

For the purpose of limit confidence interval it is allowed $n_{im} = n = 1$ and $t_{95,df_{im}} = t_{95,df}$ in **Equation 7:**

$$t_{95,df\,im} \cdot (s_{im} + s \cdot \sqrt{2}) \leq p \cdot GVE \ (8)$$

2.3. Monitoring emissions by indicative surrogate parameters and by combining predicative and indicative parameters

Indicative surrogate parameters are those process parameters for the work of emission treatment devices (Brinkmann et al. 2018). It is proved convenient for such parameters instead of concentrations to model concentration fall of emissions ($\widehat{\Delta y}_{1/2\,hr}$) and as validated values:

$$V_{1/2\,hr} = \widehat{\Delta y}_{1/2\,hr} - t_{95,df} \cdot s \cdot \sqrt{\frac{n+1}{n}} \quad (9)$$

Symbol $\widehat{\Delta y}_{1/2\ hr}$ is model for concentration fall in emission treatment and as such it is always negative, where $\widehat{\Delta y}_{1/2\ hr}$ is indicative model for concentration fall (30 min average). Confidence interval limitation, **Equation 2** for indicative surrogates is not required.

For indicative surrogate it is also possible to limit concentration fall values like limiting values (GVE) for emission concentrations and use it in validation of results with some developed logical methods for combination consisting of more indicative models

Under considerations are combinations of predictive and indicative surrogate parameter models for situation as on **Figure 1**.

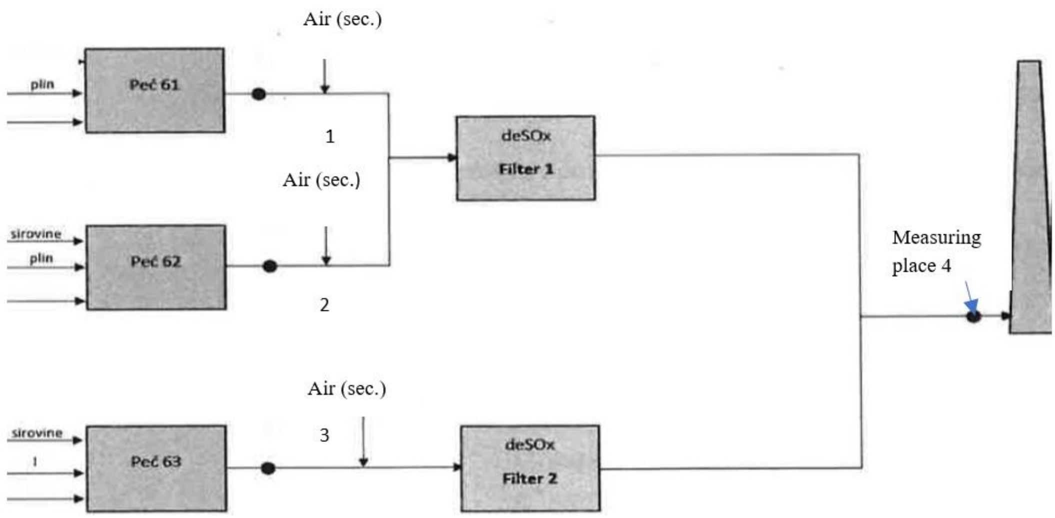

Figure 1. Combined models of predictive and surrogate parameters for emission monitoring when deSOx emission treatment is used (in glass manufacturing installation).

It uses predictive regression and indicative mass balance models for emission flows of SOx, **Figure 1** and for case of combination of two deSOx devices with calculated emission comparison for measuring place 4 (stack). The emissions are designed from furnaces 61, 62, 63, with their own developed (regression) models and with serial/parallel connections of deSOx using indicative models. The validated result of the model (30 min average) is:

$$V_{1/2\ hr,A} = D_1\left(\hat{y}_{1/2\ hr,1} - t_{95,df\ M-2}\cdot\sqrt{\frac{n+1}{n}{s^2}_{r1}}\right) + D_2\left(\hat{y}_{\frac{1}{2}hr,2} - t_{95,df\ M-2}\cdot\sqrt{\frac{n+1}{n}{s^2}_{r2}}\right) + (D_1+D_2)\left(\widehat{\Delta y}_{\frac{1}{2}hr,1} - t_{95,df\ M-2}\cdot\sqrt{\frac{n+1}{n}{s^2}_{i1}}\right) + D_3\left(\hat{y}_{\frac{1}{2}hr,3} + \widehat{\Delta y}_{1/2\ hr,2} - t_{95,df\ M-2}\cdot\sqrt{\frac{n+1}{n}{s^2}_{r3}} - t_{95,df\ M-2}\cdot\sqrt{\frac{n+1}{n}{s^2}_{i2}}\right) \qquad (10)$$

where $\hat{y}_{1/2\ hr,1}$, $\hat{y}_{1/2\ hr,2}$, $\hat{y}_{1/2\ hr,3}$ are 30 min average concentrations calculated by regression models $_{r1,\ r2,\ r3}$ for furnaces 61, 62, 63 (***Figure 1),*** $\widehat{\Delta y}_{1/2\ hr,1}$, $\widehat{\Delta y}_{1/2\ hr,2}$ 30 min average of concentration fall of pollutant achieved on deSO$_X$ devices 1 and 2 and obtained by mass balance models of indicative type $_{i1}$, $_{i2}$, D_1, D_2, D_3 ratio of waste gases flows from furnaces 61, 62 i 63 , A aggregation of the models ($V_{1/2\ hr,A}$). (In this case the correction of validated value for oxygen level of waste gas is omitted because it is not a part of the model).

In recent model used in glass manufacturing, equation of type **Equation 10** was used for construction of more developed combined model directly using estimation of standard deviation from measurement on place 4 beyond the stack and applying some mathematical techniques for maximization using ratio of waste gases flows, D_1, D_2, D_3 in limitation of confidence level for the model.

The sensitivity analysis for surrogate parameters in models should also be the part of model construction and later part for correction of the models as statistical learning (Smith&Smith 2007). Some procedures for the task were already developed.

2.4. Types of models for surrogate parameters already proposed

According to permitting system the proposal for models is given to authorities. Among predictive and indicative models now proposed for use are linear regression models and some mass balance models and artificial neural network models. Still missing other models of balance types: energy and process balances models.

Regression models are characterized by more parameters than other models and as such are convenient for modelling using predictive (process) parameters according to results of sensitivity analysis. Artificial neural network models proposed for now have only two input (surrogate) parameters and its potential for more surrogate parameters is not yet, according to that fact fully utilized. The same could be told for the low number of neuron layers now used in networks. The data about the potential of that models are still missing but their advantage over regression models in future could also be that they are nonlinear, using various types of modelling nonlinear functions (Mesellem et al. 2021). Mass balance models are used mostly for indicative parameters so the problem of number of parameters and their statistical significance in model is not yet so crucial.

By these types of models, the choice of models is not exhausted. Some approaches, even changes the basic statistics (frequentist or Bayesian) could be considered. For example, the models on Bayesian statistics, which are statistically fully functional according to used parameters and confidence level offers the new ways of optimizations of models using Bayes theorem (Sivia 2002). The other types of the models also exist which monitoring on surrogate parameters could apply (Tahraoui et al. 2021).

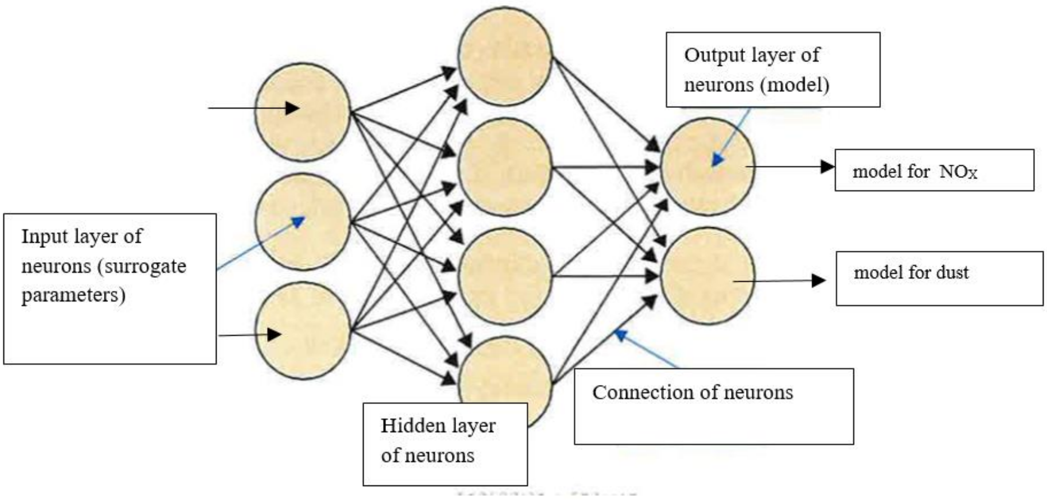

Figure 2. Model of artificial neural network with one active hidden layer for monitoring emissions of NOx and dust.

2.5. Correction of the models, statistical learning and confidence interval checking

The conditions for surrogate parameters monitoring must hold correction of the models according to the results from direct measurement in later period and for these statistical methods (statistical learning) are considered for all types of model already proposed (Hastie et al 2001). Correction of the models could be mathematically the same procedure as developing new models (Mendenhalll&Sinchic1988; Rozendaal 1999), but it seems convenient to use techniques of optimization of existing models. Now exists optimization procedures for regression and mass balance models (Press et al 2002) and for artificial neural network models (Kelleher 2019) but also the broader framework, for optimization for some other types of models still not proposed is already established. The main difference of procedures exists between neural artificial network and other types of models.

For regression models corrected value of estimation of standard deviation of population is:

$$s^2 = \frac{SSE}{n-(k+1)} \quad (11)$$

where *SSE* is calculated as:

$$SSE = \boldsymbol{Y'Y} - \boldsymbol{\hat{\beta}'X'Y} \qquad (12)$$

$\boldsymbol{Y}$ are results of regression model (vector), $\boldsymbol{X}$ membership (surrogate parameters) matrix for regression model, $\boldsymbol{\hat{\beta}}$ coefficients of regression model (vector), SSE sum of square errors of the model, *n* number of samples, k number of parameters (Mendenhall & Sinchich 1988).

For mass balance models, both indicative and predictive after the correction of the models the estimation of standard deviation is after correction:

$$s = \sqrt{\frac{\sum_{i=1}^{M}(e_i-\bar{e})^2}{M-2}} \quad (13)$$

where:
- for predictive surrogates: $e_i = y_i - \hat{y}_i$ (14),
- for indicative surrogates: $e_i = \Delta y_i - \widehat{\Delta y}_i$ (15)

and

$$\bar{e} = \frac{\sum_i e_i}{M} \quad (16),$$

$\hat{y}_i$ are calculated values of the predictive model, y_i measured concentration values, Δy_i measured concentration fall obtained as $E_{mj,2} - E_{mj,1}$, $\widehat{\Delta y}_i$ calculated values of indicative model, i pair index of measured-calculated values, M number of measured-calculated pairs, $df = M - 2$ degrees of freedom for model confidence interval, e_i measured-calculated pair error, all obtained after correction of the model.

The learning function as:

$$L = \frac{1}{M} \cdot \sum_{i=1}^{M} [y_i - \hat{y}_i]^2 \quad (17)$$

is used for the optimization of artificial neural networks.

Combined models mentioned before theoretically needs correction both for predictive and indicative models and combined models itself. One way of solving the problem is learning on surrogates' models but testing only the combined model. Limit confidence interval could be checked from **Equations (7)** and **(8)** but it is also possible to do this through control parallel measurement using reference methods, for those pollutants not covered by confidence interval limit, through variability test as:

$$s_D \leq \sigma_0 \cdot k_v \quad (18)$$

where:

$$\sigma_0 = \mu \cdot GVE/1{,}96 \quad (19)$$

Equation for model limit confidence interval considering direct measurement is:

$$1{,}96 \cdot (s_D + s \cdot \sqrt{2}) \leq \mu \cdot k_v \cdot GVE \; (20)$$

where σ_0 are determined standard deviation estimation from variability test **Equation 19,** s_D standard deviation estimation of differences from the variability test, k_v coefficient from in-measuring table, μ relative measurement uncertainty appropriate for the control measurement.

2.6. Some examples of surrogate parameters

Examples considering waste incineration/co-incineration cases could not be given yet. But examples from high temperature technologies in some way alike, give some ideas for surrogate parameters for waste incineration/co-incineration.

Some surrogate parameters from those technologies are:

- **in glass manufacturing** (waiting approval):

Predictive surrogate parameters: secondary air flow, ratio of gas and secondary air flows, rate of changes ratio of gas and secondary air flows in furnaces, temperature of air for combustion in furnaces.
Indicative surrogate parameters: flow of deSOx agents, devices efficiency factor, mass balance in filter
Qualitative surrogate parameters: glass recipes - colour of raw materials (dimensionless characteristics). (The qualitative surrogates are used for correction of predictive surrogates in more elaborate mass balance models.)

- **in mineral wool production** (in real use):

Predictive surrogate parameters: the raw materials input, air flow in furnaces, air temperature, amount of oxygen, volume ratio of O_2 and CO in waste gas, waste gas temperature, content of raw material and fuel, input and output temperatures of cooling water, chemical content of raw materials and fuel, chemical content of melt for wool production.
Indicative surrogate parameters: pressure in filters, temperature in filters, volume ratio of waste gas after waste gas treatment device.

- **for large combustion plants** (waiting approval):

only predictive surrogate parameters: flow of natural gas, volume ratio of oxygen in waste gas, flow of produced steam.

3. CONCLUSIONS

It is not an easy task to prepare administrative and technical frame for surrogate parameters model emission air monitoring, but a lot of work is already done. The questions of model types for monitoring and complicated nomenclature associated, with unified symbols for models $\hat{y}$ and $\widehat{\Delta y}$ and other issues could be considered finished for the purpose of administrative procedures and for use for regulation purposes in permits. The procedures and basic rules for statistical learning, testing and correction of the models are also defined and for cases of using indicative parameters also some issues of validation of results of modelling.

Even as examples considering waste incineration/co-incineration cases could not be given yet, examples of model proposals and parameters from the other high temperature technologies, some waiting approval and some from real use, could give ideas for waste incineration/co-incineration surrogate parameters modelling. Combined models one could propose for solution for complicated situation of monitoring, where the simple (elemental) models are not sufficient.

One always must consider that proposals for models are always coming from installations (operators) and that prepared procedures must treat them accordingly. So, the administrative procedures should be opened for proposals of users, which from the other side could improve the existing administrative procedures and technical issues.

The use of surrogate parameters for other purpose, supporting direct measurement (monitoring) is under further research because of always existing importance of confirmation results of direct measurement monitoring.

4. REFERENCES

Brinkmann T et al. (2018) JRC Reference report on monitoring of emissions to air and water from IED installations. Industrial emissions directive 2010/75/EU, EUR 29261. Aviable on: http://www.eippcb.jrc.ec.europa.eu, Cited 15 Dec 2023

Hastie T, Tibshirani R, Friedmann J (2001) Statistical learning. Springer, New York

Kelleher JD (2021) Deep learning (in croatian). Mate d.o.o., Zagreb

Mendenhall W, Sinchich T (1998) Statistics for the engineering and computer sciences. Dellen publishing company, San Francisco

Mesellem Y et al., Kem. Ind. 70 (1-2) (2021) 1-12

Press WH, Teukolsky SA, Vetterling WT, Flannery BP (2002) Numerical recipies in C++ - the art of scientific computing, 2nd ed. Cambridge unversity press, Cambridge

Rozendaal M (1999) Impact of coal quality on NOx emissions from power plants. Delft University of technology, Delft

Rumenjak D (2021) Predictive surrogate parameters (PEMS) as a way of monitoring emissions from industrial plant (in croatian). Kem. Ind. 70 (9-10) 621–622

Sivia DS (2002) Data analysis, A Bayesian tutorial. Oxford university press, New York

Smith J, Smith P (2007) Environmental modelling – an introduction. Oxford university press, New York

Tahraoui H et al., Kem. Ind. 70 (1-2) (2021) 675-691