EGU21-16027, updated on 27 Sep 2021
https://doi.org/10.5194/egusphere-egu21-16027
EGU General Assembly 2021

Reconstruction of high frequency methane peaks from measurements of metal oxide low-cost sensors using machine learning

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Deploying a dense network of sensors around emitting industrial facilities allows to detect and quantify possible CH_4 leaks and monitor the emissions continuously. Designing such a monitoring network with highly precise instruments is limited by the elevated cost of instruments, requirements of power consumption and maintenance. Low cost and low power metal oxide sensor could come handy to be an alternative to deploy this kind of network at a fraction of the cost with satisfactory quality of measurements for such applications.

Recent studies have tested Metal Oxide Sensors (MO_x) on natural and controlled conditions to measure atmospheric methane concentrations and showed a fair agreement with high precision instruments, such as those from Cavity Ring Down Spectrometers (CRDS). Such results open perspectives regarding the potential of MOx to be employed as an alternative to measure and quantify CH_4 emissions on industrial facilities. However, such sensors are known to drift with time, to be highly sensitive to water vapor mole fraction, have a poor selectivity with several known cross-sensitivities to other species and present significant sensitivity environmental factors like temperature and pressure. Different approaches for the derivation of CH_4 mole fractions from the MO_x signal and ancillary parameter measurements have been employed to overcome these problems, from traditional approaches like linear or multilinear regressions to machine learning (ANN, SVM or Random Forest).

Most studies were focused on the derivation of ambient CH_4 concentrations under different conditions, but few tests assessed the performance of these sensors to capture CH_4 variations at high frequency, with peaks of elevated concentrations, which corresponds well with the signal observed from point sources in industrial sites presenting leakage and isolated methane emission. We conducted a continuous controlled experiment over four months (from November 2019 to

February 2020) in which three types of MOx Sensors from Figaro® measured high frequency CH_4 peaks with concentrations varying between atmospheric background levels up to 24 ppm at LSCE, Saclay, France. We develop a calibration strategy including a two-step baseline correction and compared different approaches to reconstruct CH_4 spikes such as linear, multilinear and polynomial regression, and ANN and random forest algorithms. We found that baseline correction in the pre-processing stage improved the reconstruction of CH_4 concentrations in the spikes. The random forest models performed better than other methods achieving a mean RMSE = 0.25 ppm when reconstructing peaks amplitude over windows of 4 days. In addition, we conducted tests to determine the minimum amount of data required to train successful models for predicting CH_4 spikes, and the needed frequency of re-calibration / re-training under these controlled circumstances. We concluded that for a target RMSE <= 0.3 ppm at a measurement frequency of 5s, 4 days of training are required, and a recalibration / re-training is recommended every 30 days.

Our study presents a new approach to process and reconstruct observations from low cost CH_4 sensors and highlights its potential to quantify high concentration releases in industrial facilities.