Reply to the comments of Anonymous Referee #1 on the manuscript "Long-term behavior and stability of calibration models for NO and NO$_2$ low cost sensors"

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We sincerely thank the reviewer for the positive review and the constructive feedback to improve the manuscript. Please find the detailed replies to all points made by the reviewer. In the following, the reviewer's comments are given in black and the replies in blue and the revised text in green.

This manuscript evaluated NO and NO$_2$ sensors' performance during long-term deployments. Calibration models and evaluation metrics are described in detail, supporting relative conclusions. The manuscript is organized well, and this topic is important for the field deployment of air quality sensors. Therefore, I would recommend accepting the manuscript after minor revision.

1. It is good to use the Taylor diagram to show multiple metrics. It will be helpful to describe where an ideal sensor should locate in the Taylor diagram.

Reply: An ideal model/sensor would have Pearson's correlation coefficients (r) = 1 and normalised standard deviation $\sigma_{\hat{y}}/\sigma_y$ = 1. It would be located at the point marked as Ref in Figure 5. We already described the location of this reference in the caption, but reworded to clarify the meaning of the reference point.
The reference point gives the location of a perfect model/sensor.

2. On Page 5, please explain more about equation 1. It is unclear why the author would like to address relative humidity in this form. In addition, more information is needed regarding the importance of Δt_0.

Reply: First and foremost, there was a typo in the equation. Previously the exponential term was $exp(-\frac{\Delta t}{\Delta t_0})$, but the exponent should have been without minus (-). We now corrected the equation in the revised paper as below:

$$D_{RH} = \sum_{\Delta t=0}^{-500} \Delta S_{RH}(t + \Delta t) * exp(\frac{\Delta t}{\Delta t_0}). \tag{1}$$

As mentioned in Section 2.3, Equation (1) was investigated in the earlier study (Mueller et al.: Design of an ozone and nitrogen dioxide sensor unit) which utilized the same NO_2 sensor model, Alphasense NO2-B43F. In the laboratory testing of the study, it was observed that the amplitude of the NO_2 sensor response, caused by changes of relative humidity (RH), was similar to the magnitude and the rate of RH variation (Figure 2 in Mueller et al.). In addition, it was found in this study that the NO_2 sensor showed a delayed and exponentially decaying response upon changes in relative humidity. The term D_{RH} was introduced for an approximation of this behavior and including D_{RH} as a predictor variable in the calibration models largely mitigates this memory effect. In addition, the earlier study demonstrated that the model with D_{RH} had lower RMSE than the model without it. In the equation, $\sum_{\Delta t=0}^{-500} \Delta S_{RH}(t+\Delta t)$ signifies the summation of RH changes in the past 500 minutes, and since the effect of RH variation exponentially decreases over time and returns to zero, the exponential term $exp(\frac{\Delta t}{\Delta t_0})$ was chosen. Various values of Δt_0 were examined during the selection of calibration models because as demonstrated in Mueller et al., RH changes in the field measurement would differ from those in the laboratory test, and the precise physical cause of this signal is unknown. The explanations in Section 2.3. have been extended and read now as follows:

In an earlier study by Mueller et al. (2017), it was observed that for the NO_2 sensors the amplitude of the sensor response caused by varying relative humidity is of similar magnitude than the sensor response caused by typical ambient levels of NO_2. In addition, it was found in Mueller et al. (2017) that the NO_2 sensors showed a delayed and exponentially decaying response upon changes in relative humidity. Therefore, an additional variable, D_{RH}, was introduced for compensation of the effect of changing relative humidity on the raw sensor signal.

$$D_{RH} = \sum_{\Delta t=0}^{-500} \Delta S_{RH}(t+\Delta t) * exp(\frac{\Delta t}{\Delta t_0}). \tag{2}$$

ΔS_{RH} represents the change in relative humidity (in %), Δt is the corresponding time lag in minutes and Δt_0 is a time constant. Changes in relative humidity up to 500 minutes back in time are considered and weighted using the exponential term $exp(\frac{\Delta t}{\Delta t_0})$. Similar to Mueller et al. (2017), various values for Δt_0 were examined in this study (60, 90, 120, and 150 minutes) for finding the value that leads to the best performing sensor calibration models.

3. On Page 19-20, the author summarized potential reasons causing the deterioration of sensors and highlighted meteorological events and relative humidity. This paper also discusses the aging of NO_2 sensors but identified ozone O_3 as the major cause (Li et al., Characterizing the Aging of Alphasense NO_2 Sensors in Long-Term Field Deployments). It will be interesting to see why different reasons for sensor aging were identified.

Reply: Thanks for pointing to this interesting paper. We observed two issues during the second colocation period. As described in the paper, the sensor systems were malfunctioning during several short time periods. As described, the exact reasons for this

remain unknown. We speculate that over time the sensor housing might have lost its watertightness and humidity might have entered into the sensors or other parts such as the electronics altering temporarily the response behavior of the sensor units. Second, and independent from these malfunctioning periods, we see that the performance of the NO_2 sensors has significantly decreased over time. In our paper we only describe the loss in data quality without speculating on the underlying reasons. The observed degradation of the sensor performance is in agreement with the findings of Li et al. (2021). Based on Li et al. (2021), it is reasonable to argue that the observed decrease can be explained by saturation of the O3 scrubber of the NO_2 sensors. Annual mean concentrations of O_3 in Haerkingen and Zuerich (urban background) are about 21 ppb and 25 ppb, respectively, meaning that the expected lifetime of the ozone scrubber is about 13 to 17 months (comparable to the situation in Pittsburgh in the Li et al. (2021) paper). We added the following text to section 2.1:

It should be pointed out here that the used NO_2 sensors have an O_3 scrubber membrane mounted on top of the inlet to prevent the interference from ambient O_3. The O_3 scrubber has reported to have a capacity of 250 ppm h of O_3 (Li et al., 2021) and thus a limited lifetime.

The following text was added to section 3.2.2:
A similar degradation of the performance of the same NO_2 sensor has been reported by Li et al. (2021). In their study, sensor performance degradation was noticeable after 200 - 400 days of deployment, a time period that was in agreement with the expected lifetime of the O_3 scrubber as calculated from its reported capacity and the O_3 concentration at the deployment site. It is therefore reasonable to assume that the decrease in NO_2 sensor performance observed in this study is also influenced or caused by saturation of the O_3 scrubber of the NO_2 sensors. At the co-location site Haerkingen and in the urban background of Zuerich, annual mean concentrations of O_3 are about 21 ppb and 25 ppb, respectively. This means that the expected lifetime of the O_3 scrubber is about 13 to 17 months, which is comparable to the situation described by Li et al. (2021).