

Temporal evolution of vehicle exhaust plumes in a congested street canyon environment

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1. Sensor calibration and QA/QC

It is important that gas sensors respond linearly over the concentration range of interest. Laboratory calibration was conducted before and after the measurements. Standard CO and CO₂ gas (100 ppm CO and 10% CO₂/N₂, Linde HKO Ltd., Hong Kong, China) concentrations were produced with a dynamic calibrator (T700U, Teledyne, Thousand Oaks, CA, USA) combined with a zero-gas generator (T701, Teledyne, Thousand Oaks, CA, USA) for CO gas and a pure N₂ source (Linde HKO Ltd., Hong Kong, China) for CO₂ gas. The NO₂ and NO were generated with a NO₂/NO/O₃ calibration source (714, 2B Technology, Boulder, CO, USA). During calibration, the gas flow rate was fixed at 1 L min⁻¹ in all cases (CO, NO₂ and NO), and validated with a flow meter (Defender 520, Mesa Labs, Lakewood, CO, USA). Prior to the experiments, both the T700U and 2B 714 instruments were allowed to warm up for 30 min. In all cases, the pollutant sensors were allowed to warm up for 3 h to ensure they had reached steady-state. Real-time direct output of pollutant concentrations and system status data were transmitted to the cloud server for online data processing and also stored locally in a memory module for backup.

Specific calibration procedure

Laboratory tests were conducted to establish the relationship of output from sensors with concentration steps of individual gases (NO₂, NO, CO, CO₂) under controlled laboratory conditions. Before the experiment, all the sensors were “warmed up” for more than 3 hours to achieve chemical equilibrium.

The sensors were exposed to each step in the calibration steps for 10 min. The steps were designed for the real-world concentration range on our monitoring site. The setting can be seen in Table S1, and there is an example of the NO step as shown in Fig.S1. Thereafter, the sensor outputs were recorded and assessed for linearity with the input concentrations. There is a linear relationship between sensor output and pollutant concentration as the Pair Differential Filter (PDF) technology offsets the impact of temperature and humidity.

Table S1. Calibration Concentration Setting

Pollutant	Concentration Setting(ppb)	Time for each step	Gas Source
NO	0,5,2,1	10min	T700U
NO ₂	0,1,0.5,0.2	10min	2B714
CO	0,4,2,1	10min	T700U
CO ₂	0,2,1,0.5	10min	T700U

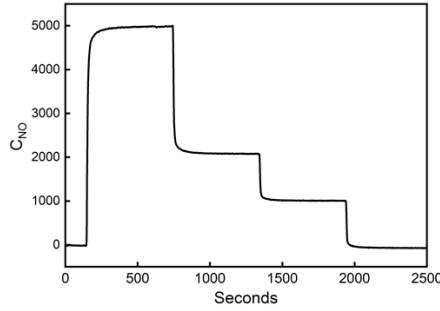


Figure S1. Calibration step of NO

Calibration instrumentation

NO₂ concentrations were produced by Model 714 NO₂/NO/O₃ Calibration Source (714, 2B Technology, Boulder, CO, USA). NO is provided by disposable 8 or 16 oz cartridges typically used for making whipped cream. Calibrated concentrations of NO₂ are produced by gas-phase titration (GPT) of NO with O₃ using the same method as conventional GPT NO/NO₂ calibrators. The concentrations of NO₂ gases are directly traceable to the NIST photometric standard for ozone – eliminating uncertainties associated with the stability of nitric oxide standards.(Birks et al., 2020)

CO₂, NO and CO concentrations were produced with a dynamic calibrator (T700U, Teledyne, Thousand Oaks, CA, USA). It includes three mass flow controllers and is capable of producing NO₂ and ozone calibrations down to 3 ppb. When generating standard gas, a zero-gas generator (T701, Teledyne, Thousand Oaks, CA, USA) for CO gas and a pure N₂ source (Linde HKO Ltd., Hong Kong, China) for CO₂ gas was combined with the T700U.

Table S2. Calibration Instrumentation

Instrument Model	Parameter	Full Scale Range	Accuracy
714, 2B Technology, Boulder, CO, USA	NO ₂	0-1000 ppb	2.0%
T700U, Teledyne, Thousand Oaks, CA, USA	CO, CO ₂ , NO	0 - 100 ppb to 0 to 10 ppm User Selectable	1.0%

2. e-folding time

No instrument responds immediately to changes in its environment. Instrument measuring a particular time-varying property $X(t)$ will not measure X exactly because the instrument takes a finite amount of time to respond to changes in X . Instead the instrument will record a measurement $X_m(t)$. The rapidity of the response of the instrument can be quantified by its time constant τ (tau), which is a measure of the time required for the instrument to respond to a sudden change in X , as shown schematically in Figure S2.

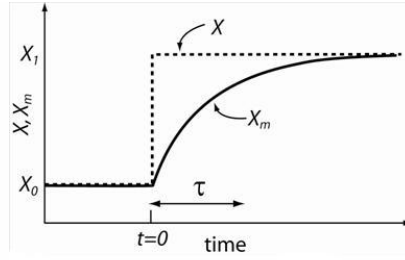


Figure S2. Schematic showing instrument response to a step change of X

When $t = \tau$ the instrument measures a value that has reduced the initial difference $X_1 - X_0$ between the true and the measured value of X by a factor of $1/e$ (≈ 0.367). Thus, τ is referred to as the e-folding time, or equivalently the time constant. Small values of τ indicate that the instrument has a rapid response. For large values of τ the instrument has a slow response.

3. Uncertainty estimation

For parameter a , We calculate a from X_3 and vehicle speed, therefore, the standard error of X_3 and the accuracy of speed sensors (0.5 m/s) can be used to estimate the indirect uncertainty of a .

To calculate the uncertainties of a , we firstly determine the partial derivatives of a with respect to X_3 and speed v :

$$\frac{\partial a}{\partial X_3} = -\frac{1}{2v} \sqrt{\frac{1}{2X_3^3}}$$

$$\frac{\partial a}{\partial v} = -\frac{1}{v^2} \sqrt{\frac{1}{2X_3}}$$

Then we calculate the uncertainties of a using the partial derivatives and the known uncertainties

$$\Delta a = \sqrt{\left(\frac{\partial a}{\partial X_3} \bigg|_{(\bar{X}_3, \bar{v})} \Delta X_3\right)^2 + \left(\frac{\partial a}{\partial v} \bigg|_{(\bar{X}_3, \bar{v})} \Delta v\right)^2}$$

Where ΔX_3 is the standard error of X_3 and Δv is the uncertainty of v .

By substituting the values of $(\partial a / \partial X_3)$, $(\partial a / \partial v)$, ΔX_3 , and Δv into the equation, we can estimate the uncertainties of a .

Note: This method assumes that the uncertainties in X_3 and v are independent and that the errors follow a normal distribution. The process of estimating the uncertainty of Q follows a similar approach, which is not presented here for brevity.

There is no standardized method to estimate the uncertainty of the deconvolution technique, we visualize the absolute error by comparing the deconvoluted data with real-world stimulus in lab experiments, as shown by the Figure S3.

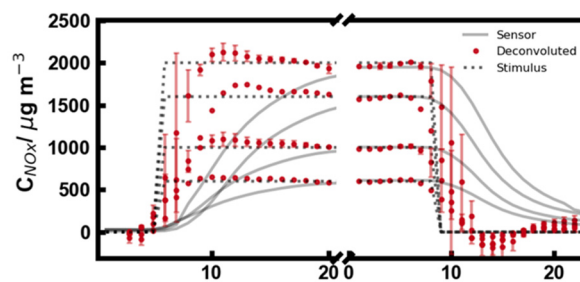


Figure S3. Imposed changes in NO_x concentration in laboratory experiments (gray dotted line), concentration measurements as gray lines and deconvoluted concentrations as red dots with error bar represent the absolute errors.

Reference

Birks, J. W., Turnipseed, A. A., Andersen, P. C., Williford, C. J., Strunk, S., Carpenter, B., & Ennis, C. A. (2020). Portable calibrator for NO based on the photolysis of N₂O and a combined NO₂/NO/O₃ source for field calibrations of air pollution monitors. *Atmospheric Measurement Techniques*, 13(2), 1001–1018.
<https://doi.org/10.5194/AMT-13-1001-2020>