Quantifying nitrogen oxides and ammonia via frequency modulation in gas sensors

Master Thesis defense seminar

Marcos F Mourão

June 1^{st} 2021

Outline

But first...

A few seconds of gratitude

- Annika Tillander - Internal Supervisor
- Mike Andersson - External Supervisor
- José M. Peña - Examiner
- Oleg Sysoev - Course Leader
- Samia Noreen Butt - Opponent

Thank you!

Problem in a nutshell

Motivation

[1]Image source: ENVIS Centre on Plants and Pollution

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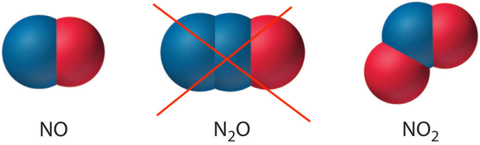
NO_x[1]:



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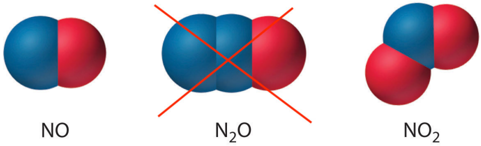

- NO_x are detrimental to the environment and humans.

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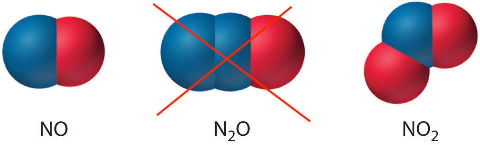

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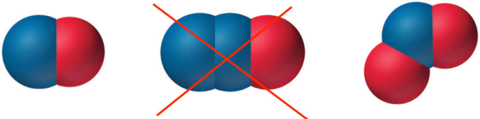

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- Ammonia can "neutralize" NO_x; producing water (H_2O) and nitrogen gas (N_2). Both harmless! - Selective catalytic reduction (SCR).

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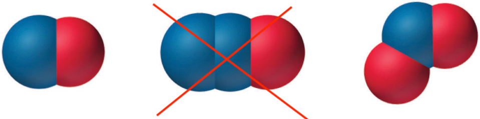

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- Ammonia can "neutralize" NO_x; producing water (H_2O) and nitrogen gas (N_2). Both harmless! - Selective catalytic reduction (SCR).
- But ammonia is also hazardous to the environment/humans.

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- **Frequency cycling**.

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1. Can frequency modulation be used to simultaneously quantify NO_x and Ammonia concentrations?
2. Does the quality of fit vary over different prediction models?

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- Shape-defining features: **Slopes and Averages**.
- Two important definitions:
  1. Mixture: an **unique** combination of gases, i.e. **no repetitions**.
  2. Exposure: a combination of gasses **with repetition**.

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Data collection

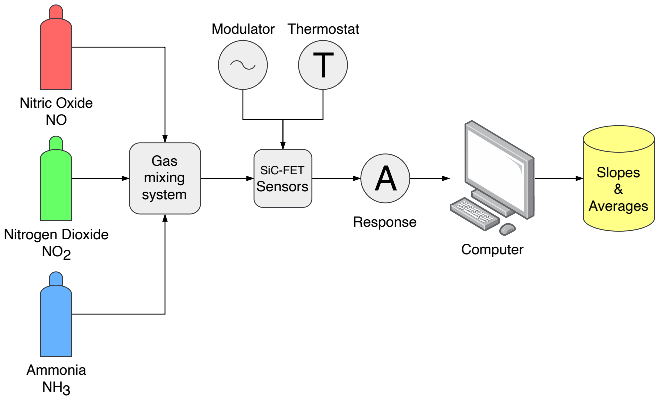

Figure: Schema of the data acquisition process.

Data

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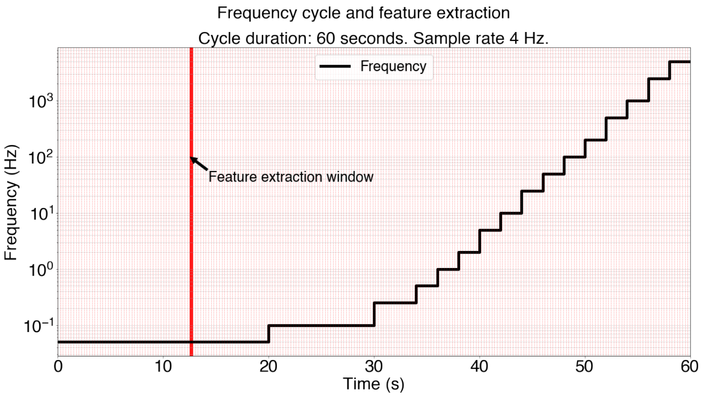

Figure: Feature measurements times per cycle. The width of the red line indicates the duration of one of the feature measurement windows as an example.

Data

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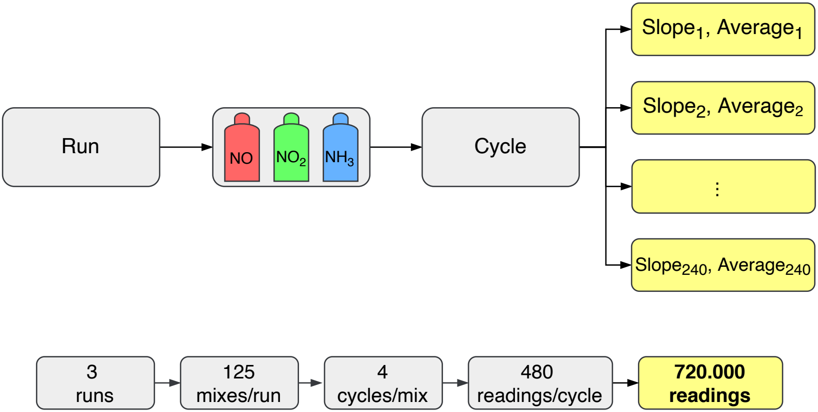

Figure: A visualization of the feature measurement process.

Data

Raw data

Row-based

Table: Sample of raw data.

Index	Exposure nr	Cycle nr	Sample nr	NO [ppm]	NO2 [ppm]	NH3 [ppm]	Freq [Hz]	Slope sensor 1 [μA/s]	Slope sensor 2 [μA/s]	Average sensor 1 [μA]	Average sensor 2 [μA]	Sensor temperature [C]
0	1	1	1	10	5	20	0.05	-18.855169	-22.588416	32.926184	27.961554	274.994683
1	1	1	2	10	5	20	0.05	-28.289268	-28.185027	25.853867	20.915297	274.980487
2	1	1	3	10	5	20	0.05	-0.390916	-0.482129	25.756138	20.794765	274.985895
3	1	1	4	10	5	20	0.05	-0.234549	-0.156366	25.697501	20.755673	275.020372
4	1	1	5	10	5	20	0.05	-0.143336	-0.247580	25.661667	20.693778	275.014964
							⋮					
100000	105	1	161	5	5	40	5.0	-38.366212	-48.495271	30.241896	24.821197	275.021724
100001	105	1	162	5	5	40	5.0	6.619507	8.521964	31.896773	26.951688	274.999415
100002	105	1	163	5	5	40	5.0	-1.941549	6.580416	31.411386	28.596792	275.011584
100003	105	1	164	5	5	40	5.0	27.401023	22.012900	38.261641	34.100017	275.009894
100004	105	1	165	5	5	40	5.0	-27.016623	-28.439121	31.507486	26.990236	275.014400
							⋮					
359995	375	4	236	20	80	5	5000.0	-0.136821	-0.158538	34.129879	30.345597	275.002007
359996	375	4	237	20	80	5	5000.0	0.010859	0.010859	34.132593	30.348312	274.986797
359997	375	4	238	20	80	5	5000.0	-0.043435	0.030405	34.121734	30.355913	274.979811
359998	375	4	239	20	80	5	5000.0	-0.117275	-0.026061	34.092416	30.349398	274.984543
359999	375	4	240	20	80	5	5000.0	0.073840	0.039092	34.110876	30.359171	274.998063

Data

Preprocessing

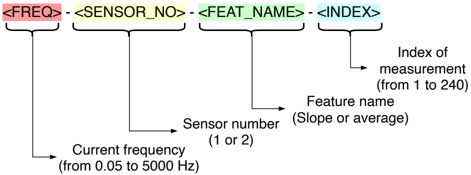

Figure: Feature naming convention.

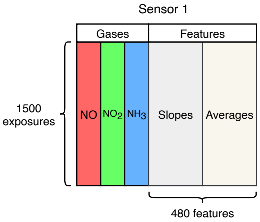

Figure: Pre-processed data structure.

Data

Preprocessed data

Column-based.

Table: Sample of pre-processed data.

Index	EXPOSURE	NO	NO2	NH3	0.05-1-slope-0	0.05-1-slope-1	...	5000.0-1-slope-239	0.05-1-avg-0	0.05-1-avg-1	0.05-1-avg-2	...	5000.0-1-avg-238	5000.0-1-avg-239
0	1.0	10.0	5.0	20.0	-18.855169	-28.289268	...	0.019546	32.926184	25.853867	25.756138	...	35.840135	35.845021
1	1.0	10.0	5.0	20.0	-28.979886	-9.251672	...	-0.056466	28.600050	26.287132	26.225237	...	35.884113	35.869996
2	1.0	10.0	5.0	20.0	-25.431240	-12.874158	...	-0.052122	29.512187	26.293647	26.238267	...	35.913432	35.900401
3	1.0	10.0	5.0	20.0	-30.126572	-8.196200	...	-0.156366	28.368758	26.319708	26.254555	...	35.939493	35.900401
4	2.0	20.0	40.0	40.0	-19.506695	-27.051368	...	-0.078183	33.180279	26.417437	26.303420	...	35.685397	35.665852
							⋮							
700	176.0	40.0	20.0	40.0	-21.011721	-25.822155	...	-0.071668	31.458621	25.003082	24.902639	...	34.554999	34.537082
701	176.0	40.0	20.0	40.0	-27.505265	-10.847911	...	0.086870	27.660766	24.948788	24.918927	...	34.504506	34.526224
702	176.0	40.0	20.0	40.0	-27.516124	-10.750182	...	-0.097729	27.647193	24.959647	24.928700	...	34.531653	34.507221
703	176.0	40.0	20.0	40.0	-27.364102	-10.875058	...	0.086870	27.666195	24.947431	24.935215	...	34.537082	34.558800
704	177.0	80.0	40.0	40.0	-20.794546	-26.195696	...	0.041263	31.640505	25.091581	25.088324	...	34.695078	34.705393
							⋮							
1495	374.0	80.0	80.0	40.0	-27.937445	-10.891346	...	-0.097729	27.166692	24.443855	24.392276	...	34.151596	34.127164
1496	375.0	20.0	80.0	5.0	-24.358394	-22.933723	...	-0.008687	30.315735	24.582305	24.530726	...	34.134765	34.132593
1497	375.0	20.0	80.0	5.0	-28.862612	-9.827186	...	-0.112931	26.916940	24.460144	24.410736	...	34.159740	34.131507
1498	375.0	20.0	80.0	5.0	-25.839531	-12.780772	...	-0.021718	27.671625	24.476432	24.430282	...	34.143452	34.138023
1499	375.0	20.0	80.0	5.0	-28.002598	-10.645937	...	0.073840	27.137373	24.475889	24.424853	...	34.092416	34.110876

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Feature averaging

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- 1500 exposures - hard to visualize.

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- Averaging all **exposures** of a particular **mixture**.
- 125 mixtures - easier (or less hard) to visualize

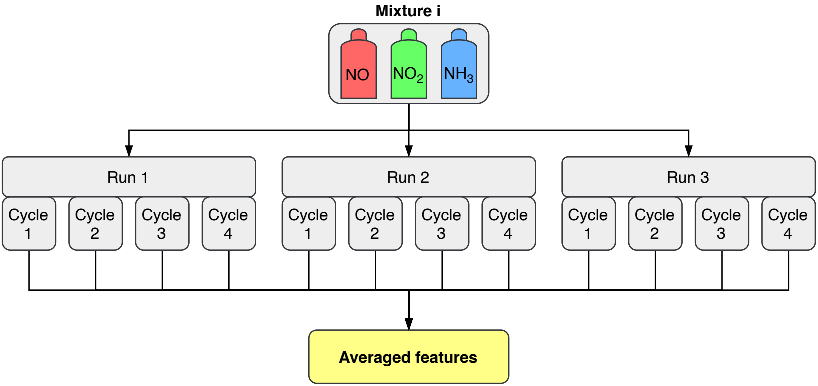

Figure: A visualization of the feature averaging process.

Data

Slopes

In each plot, each line represents a gas **mixture**.

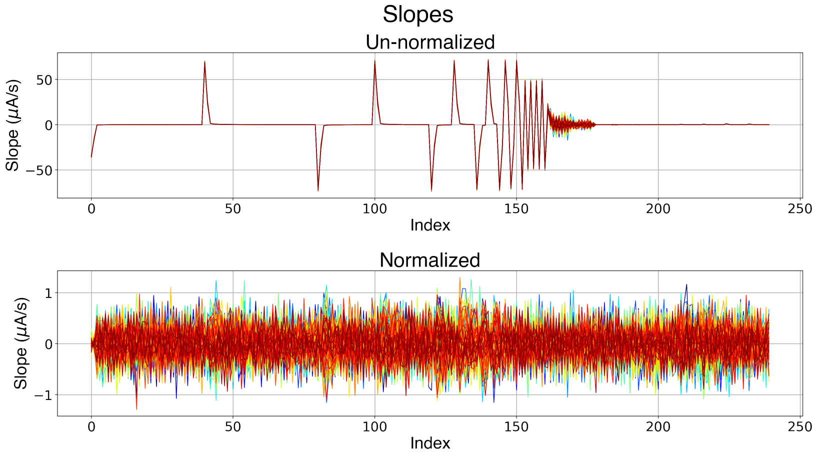

Figure: Slope features, un-normalized and normalized.

Data

Averages

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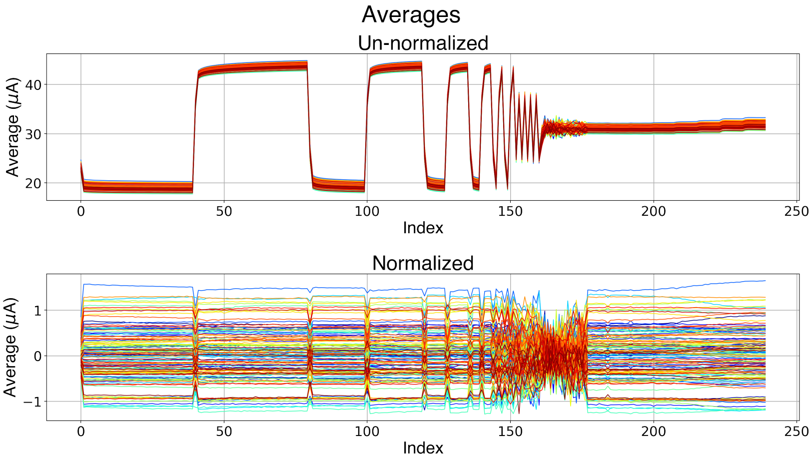

Figure: Average features, un-normalized and normalized.

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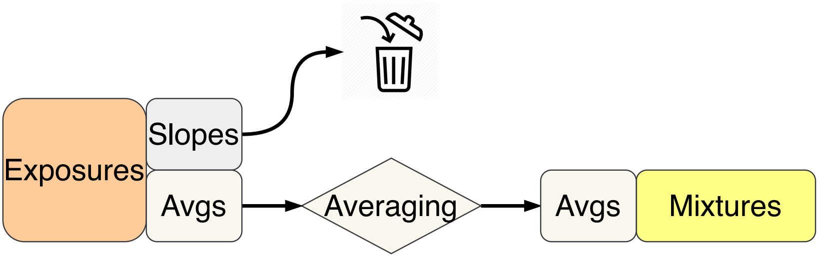
- Slope features don't change between mixtures (down to measuring noise).
- Analysis will be **also** run on averaged averages only.



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Feature averaging

Index	UNIQUE MIXTURE	NO	NO2	NH3	0.05-1-avg-0	0.05-1-avg-1	0.05-1-avg-2	...	5000.0-1-avg-238	5000.0-1-avg-239
0	0	5.0	5.0	5.0	28.983749	25.442410	25.383750	...	35.162932	35.152458
1	1	5.0	5.0	10.0	28.538652	24.933269	24.879247	...	34.622460	34.623591
2	2	5.0	5.0	20.0	29.038925	25.245278	25.181935	...	35.025637	35.023103
3	3	5.0	5.0	40.0	28.698684	25.057399	24.980686	...	34.575699	34.576649
4	4	5.0	5.0	80.0	28.738748	25.289980	25.229714	...	34.860040	34.854340
							⋮			
70	70	20.0	80.0	5.0	28.142217	24.646824	24.596195	...	34.208650	34.213536
71	71	20.0	80.0	10.0	28.615026	24.952453	24.893228	...	34.511972	34.497900
72	72	20.0	80.0	20.0	28.432463	24.705665	24.649538	...	34.317554	34.313437
73	73	20.0	80.0	40.0	28.327675	24.725143	24.685825	...	34.213989	34.197429
74	74	20.0	80.0	80.0	28.611836	25.056652	24.993128	...	34.592507	34.593548
							⋮			
120	120	80.0	80.0	5.0	28.548244	25.157684	25.103051	...	34.742313	34.749349
121	121	80.0	80.0	10.0	28.630183	25.045884	25.015773	...	34.678857	34.675690
122	122	80.0	80.0	20.0	28.420835	24.737087	24.687996	...	34.354338	34.347416
123	123	80.0	80.0	40.0	28.457189	24.743263	24.682929	...	34.327938	34.319839
124	124	80.0	80.0	80.0	28.615161	25.093255	25.046698	...	34.743829	34.734124

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- Pros: backed by literature, simple to implement.
- Cons: only linear models.

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OLS

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$$\mathbf{Y} = \mathbf{X}\mathbf{B} + \mathbf{E} \tag{1}$$

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OLS

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- The difference between the response and the prediction is the residual. The Residual Sum of Squares is defined as:

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- In this problem, features are sampled in quick succession (4 Hz). This will lead to highly correlated features $\rightarrow$ high variance of

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- While this method successfully best explains the variability in $\mathbf{X}$, the responses $\mathbf{Y}$ are not taken into account.

Methods

PLSR

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- Again, analogous to OLS, the coefficients are:

$$\widehat{\mathbf{B}}^{\mathrm{PLSR}} = (\mathbf{W}^\intercal\mathbf{W})^{-1}\mathbf{W}^\intercal\mathbf{Y} \tag{11}$$

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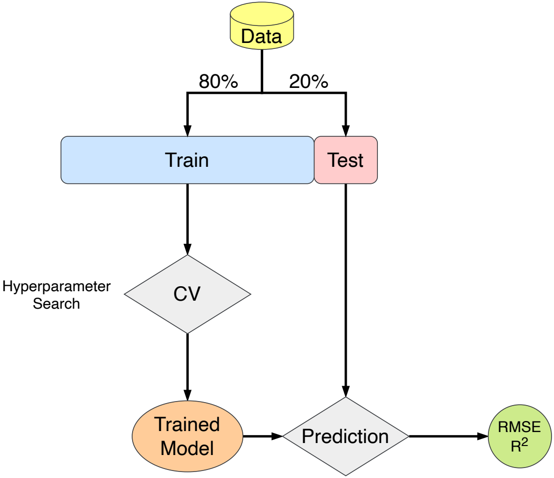
- Solving for $\widehat{\mathbf{B}}$ yields:

$$\widehat{\mathbf{B}}^{\mathrm{Ridge}} = (\mathbf{X}^{\intercal}\mathbf{X} + \boldsymbol{\lambda}\mathbf{I})^{-\mathbf{1}}\mathbf{X}^{\intercal}\mathbf{Y} \tag{14}$$

Methods



Data
80%
20%
Train
Test
Hyperparameter
Search
CV
Trained
Model
Prediction
RMSE
R2

Results

OLS

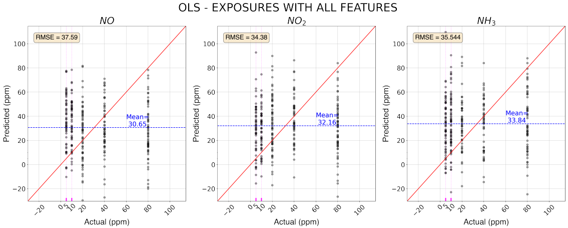


Figure: Actual vs. Predicted plot for **exposures**.

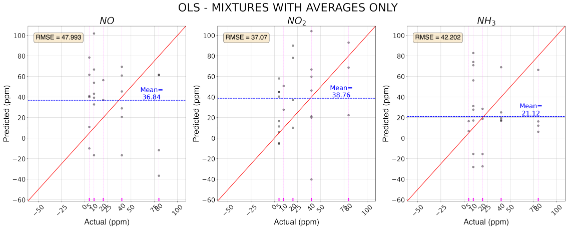


Figure: Actual vs. Predicted plot for **mixtures** - averaged average features.

Results

PCR

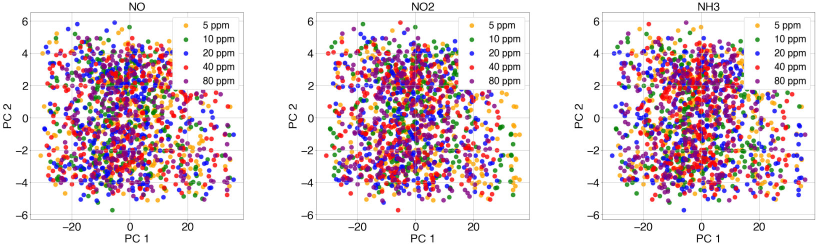
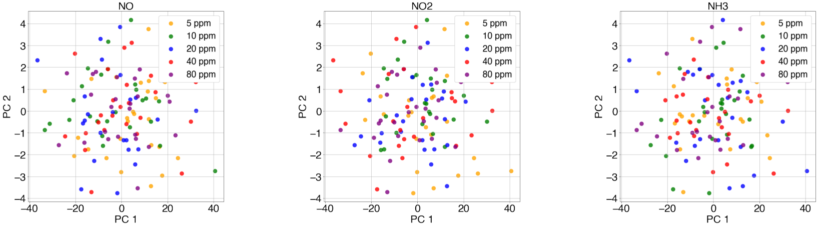


Figure: PCA for slopes and averages through **exposures**



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PCR

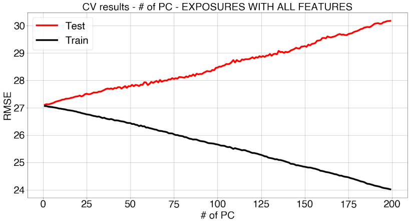
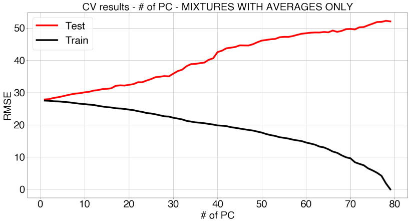


Figure: CV results for **exposures**.



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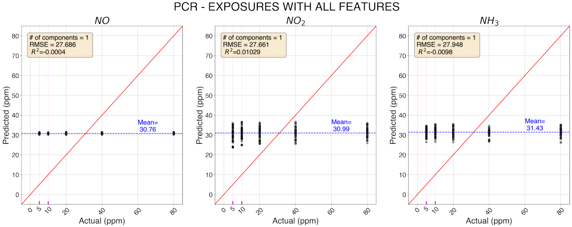
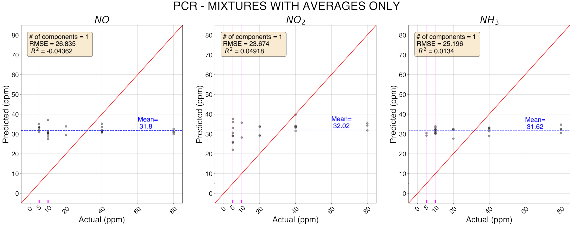


Figure: Actual vs. pred for **exposures**.



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PLSR

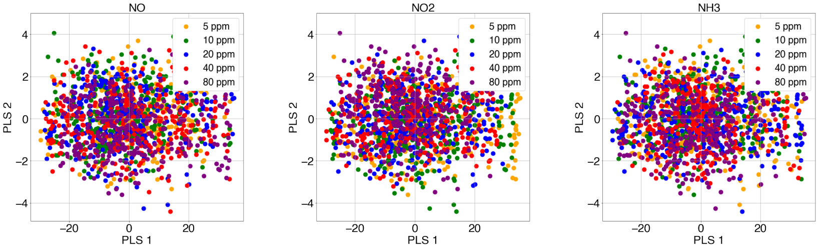
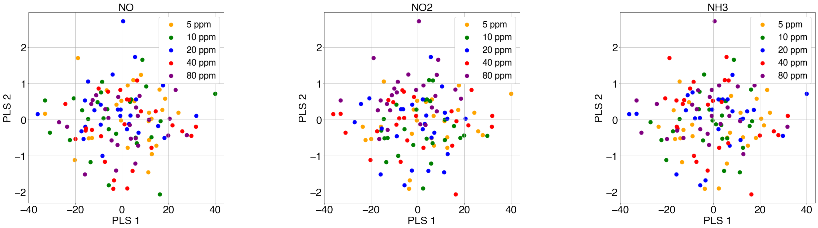


Figure: PLS scores for **exposures**.



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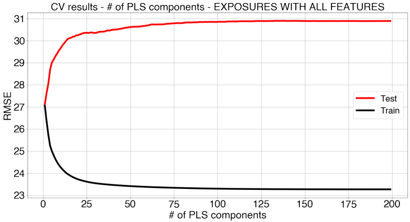

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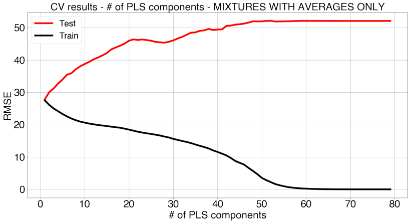

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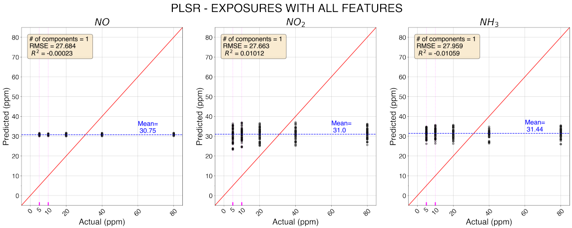

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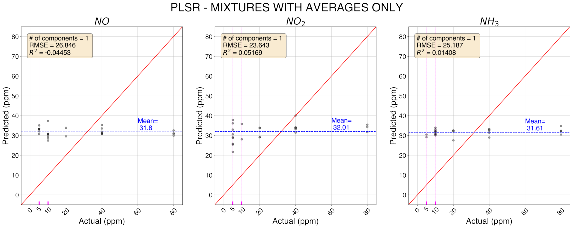

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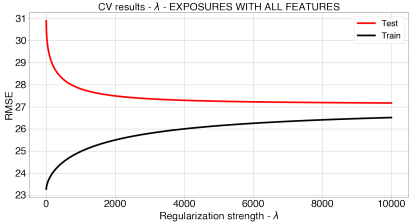
Ridge regression



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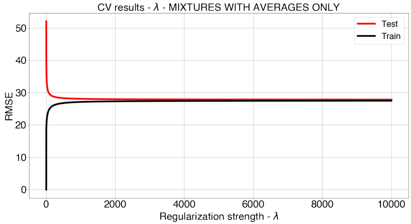

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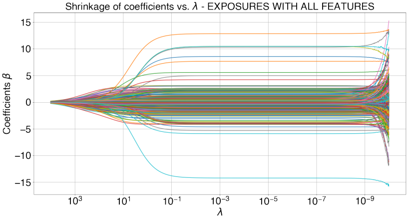
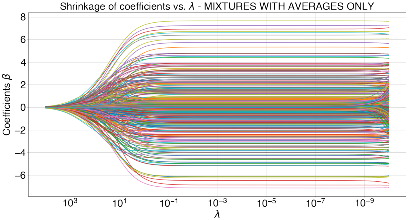


Figure: Shrinkage of coefficients for **exposures**.



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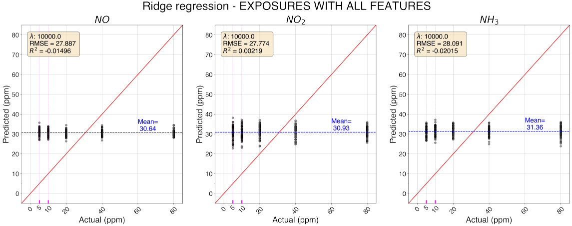

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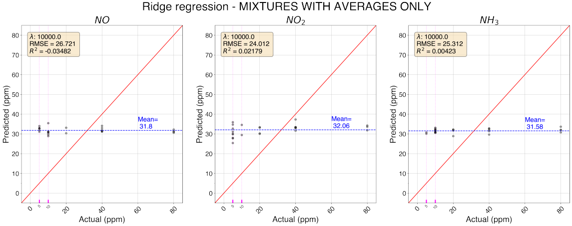

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- Results show that less complex, under-fitting models are preferred.

Discussion

Attempts at finding order

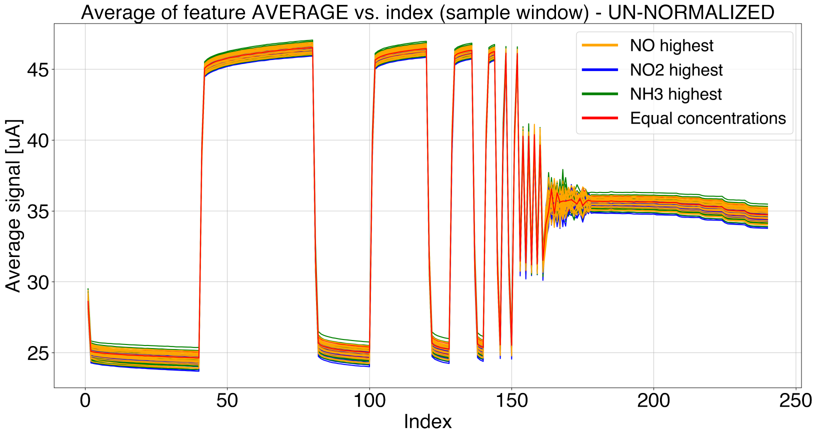

Figure: Averaged sensor average divided by predominant gas. Each line corresponds to a unique **mixture**.

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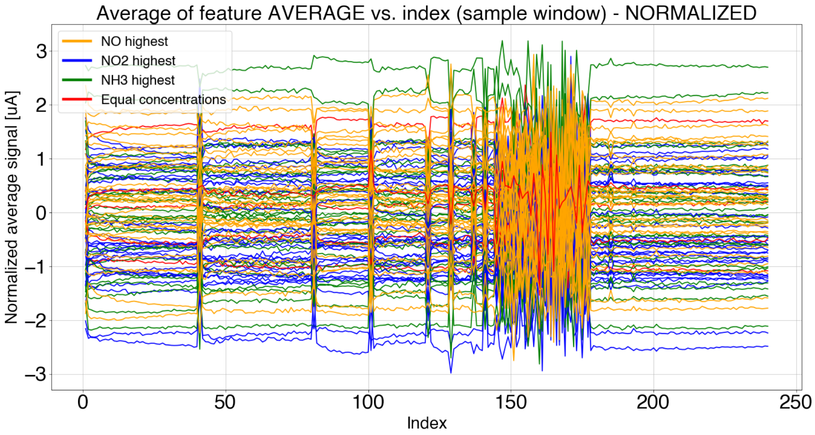

Figure: Normalized averaged sensor average divided by predominant gas. Each line corresponds to a unique **mixture**.

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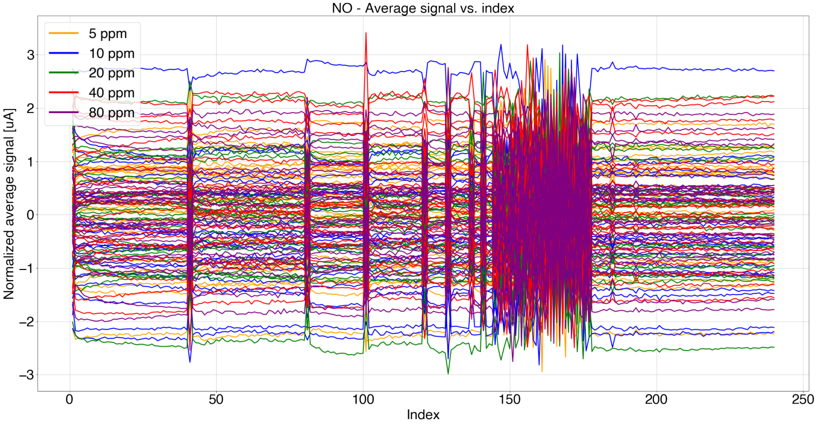

Figure: Normalized sensor averaged for NO. Each line corresponds to a unique **mixture**. The levels are the NO concentrations of the mixture.

Conclusion

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However, this work succeeded in pointing what does *not* work. Arguably, this *is as important as* know what works.

Thank you!