Blind Source Separation under the Langmuir model for chemical sensors

IEEE SAM

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Electronic nose
Principle

Electronic what?
An electronic nose = bio-inspired instrument which is able to identify and recognise Volatile Organic Compounds (VOCs) [1]
Electronic nose
Principle

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How does it work?
Electronic what?

An electronic nose = bio-inspired instrument which is able to identify and recognise Volatile Organic Compounds (VOCs) [1]

How does it work?

[Diagram showing air flow, binding, prism reaction, and chemical sensor]
Electronic what?
An electronic nose = bio-inspired instrument which is able to identify and recognise Volatile Organic Compounds (VOCs) [1]

How does it work?

For what purpose?
Biomedical, in/outdoor air monitoring, security, navigation, ...

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Electronic what?
An electronic nose = bio-inspired instrument which is able to identify and recognise Volatile Organic Compounds (VOCs) [1]

How does it work?

Chemical sensors

Air flow
Binding reaction
Prism
Chemical sensor
Light
Camera
VOC

300 μm

For what purpose?
Biomedical, in/outdoor air monitoring, security, navigation, ...
Electronic nose
Principle

Electronic what?
An electronic nose = bio-inspired instrument which is able to identify and recognise Volatile Organic Compounds (VOCs) [1]

How does it work?

![Graph showing output after subtraction over time for Propionic acid injection and cleaning.](image)

![Image showing chemical sensors with dimensions of 300 μm.](image)
Electronic what?
An electronic nose = bio-inspired instrument which is able to identify and recognise Volatile Organic Compounds (VOCs) [1]

How does it work?

[Graph showing output after reference subtraction and steady-state value for Propionic acid]

[Image of chemical sensors with a note: 300 μm]
Electronic nose
Principle

Electronic what?
An electronic nose = bio-inspired instrument which is able to identify and recognise Volatile Organic Compounds (VOCs) [1]

How does it work?

For what purpose?
Biomedical, in/outdoor air monitoring, security, navigation, ...
Electronic nose
Source Separation issue

Sensors outputs for A1

Sensors outputs for A2

Sensors outputs for the mixture

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Electronic nose
Source Separation issue

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Electronic nose
Source Separation issue

Sensors outputs for $A_1$

Sensors outputs for $A_2$

Sensors outputs for the mixture

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Electronic nose
Source Separation issue

Sensors outputs for $A_1$
Sensor 1  Sensor 2  Sensor P

Sensors outputs for $A_2$
Sensor 1  Sensor 2  Sensor P

Sensors outputs for the mixture
Sensor 1  Sensor 2  Sensor P

$C_1$

$C_2$

$\mathcal{L}$

$\mathcal{L}^{-1}$

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Physico-chemical mixture model
Chemical model

Langmuir isotherm
Langmuir isotherm

$\theta_{eq} = \frac{r_{pn} \theta}{k_{rp} c_{rn}}$

with $k_{rp}$ the affinity and $c_{rn}$ the concentration.
Physico-chemical mixture model
Chemical model

**Langmuir isotherm**

Air flow

A binding site

Prism

Free site

A\text{1}

A\text{2}

\(a_2\)

\(d_1\)

Binding reaction

\(\theta_{eq}\): fraction of occupied sites

Langmuir isotherm [2] for a multi-component gas, noting:

\[
\theta_{eq} = \frac{r_{pn}}{r_p + c_{rn}}
\]

with

\(K_r\) the affinity and

\(c_{rn}\) the concentration

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Physico-chemical mixture model
Chemical model

Langmuir isotherm

Air flow
Prism
A binding site

Occupied site

$\theta_{eq}$: fraction of occupied sites

Langmuir isotherm [2] for a multi-component gas, noting:

$\frac{r}{\text{VOC}} \text{ the VOC}$
$\frac{p}{\text{the chemical sensor}}$
$n_{\text{the mixture}}$

$\theta_{eq} = \frac{r \cdot p \cdot n}{k_{rp} \cdot c_{rn} \cdot 1}$

$k_{rp}$ the affinity and $c_{rn}$ the concentration
Langmuir isotherm

\[ \theta_{eq} = \frac{r_p n}{c_n + k_r c_n} \]

- $\theta_{eq}$: fraction of occupied sites
- $r_p$: the VOC
- $n$: the chemical sensor
- $c_n$: the mixture

with $k_r$ the affinity and $c_n$ the concentration
Langmuir isotherm

\[ \theta_{eq} = \frac{\theta_1 \cdot \theta_2}{\theta_1 + \theta_2} \]

1 chemical sensor

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Langmuir isotherm
Langmuir isotherm

\[ \theta_{eq} : \text{fraction of occupied sites} \]
Langmuir isotherm

Langmuir isotherm [2] for a multi-component gas, noting:

\[
\begin{align*}
\theta^{eq}_{rpn} &= \frac{k_{rp}c_{rn}}{1 + \sum_{r=1}^{R} k_{rp}c_{rn}} \\
\text{with } k_{rp} &\text{ the affinity and } c_{rn} \text{ the concentration}
\end{align*}
\]
Surface Plasmon Resonance model

Additivity assumption
Assuming that $e_b$ is in the nanometer range, the measure $y_{rpn}$ is proportional to the fraction of occupied sites $\theta_{rpn}^{eq}$ (with $m_r$ the mass of $A_r$) [3, 4]:

$$y_{rpn} = \gamma m_r \theta_{rpn}^{eq}$$
Assuming that $e_b$ is in the nanometer range, the measure $y_{rpn}$ is proportional to the fraction of occupied sites $\theta_{rpn}^{eq}$ (with $m_r$ the mass of $A_r$) [3, 4]:

$$y_{rpn} = \gamma m_r \theta_{rpn}^{eq}$$

Additivity assumption

The measure of the mixture is assumed to be the sum of the individual contributions:

$$y_{pn} = \sum_{r=1}^{R} y_{rpn} = \gamma \frac{\sum_{r=1}^{R} m_r k_{rp} c_{rn}}{1 + \sum_{r=1}^{R} k_{rp} c_{rn}}$$
Surf erc Plasmon Resonance model

Assuming that $e_b$ is in the nanometer range, the measure $y_{rpn}$ is proportional to the fraction of occupied sites $\theta^\text{eq}_{rpn}$ (with $m_r$ the mass of $A_r$) [3, 4]:

$$y_{rpn} = \gamma m_r \theta^\text{eq}_{rpn}$$

Additivity assumption

The measure of the mixture is assumed to be the sum of the individual contributions:

$$y_{pn} = \sum_{r=1}^{R} y_{rpn} = \gamma \frac{\sum_{r=1}^{R} m_r k_{rp} c_{rn}}{1 + \sum_{r=1}^{R} k_{rp} c_{rn}}$$
Assuming that $e_b$ is in the nanometer range, the measure $y_{rpn}$ is proportional to the fraction of occupied sites $\theta_{eq}^{rpn}$ (with $m_r$ the mass of $A_r$) [3, 4]:

$$y_{rpn} = \gamma m_r \theta_{eq}^{rpn}$$

Additivity assumption

The measure of the mixture is assumed to be the sum of the individual contributions:

$$y_{pn} = \sum_{r=1}^{R} y_{rpn} = \gamma \frac{\sum_{r=1}^{R} m_r k_{rp} c_{rn}}{1 + \sum_{r=1}^{R} k_{rp} c_{rn}}$$
Problem dimensions
Physico-chemical mixture model
Global model

Problem dimensions

R. VOCs

A1 A2 AR

...
Physico-chemical mixture model

Global model

Problem dimensions

R VOCs

A₁ A₂ Aᵣ

N experiments

Experiment 1 Experiment 2 Experiment N
**Physico-chemical mixture model**

**Global model**

---

**Problem dimensions**

R VOCs

![VOCs diagram](image)

A\_1 \ A\_2 \ \ldots \ A\_R

N experiments

![Experiments diagram](image)

Experiment 1 \ Experiment 2 \ \ldots \ \text{Experiment N}

P sensors

![Sensors diagram](image)

Sensor 1 \ Sensor 2 \ \ldots \ Sensor P
Physico-chemical mixture model
Global model

Problem dimensions

R VOCs
A_1 A_2 A_R

N experiments
Experiment 1 Experiment 2 Experiment N

P sensors
Sensor 1 Sensor 2 Prism Sensor P

chemical sensor, mixture
VOC

y_{pn} = \gamma \frac{\sum_{r=1}^{R} m_r k_{rp} c_{rn}}{1 + \sum_{r=1}^{R} k_{rp} c_{rn}}

VOC, chemical sensor
VOC, mixture

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Physico-chemical mixture model
Global model

Problem dimensions

- R VOCs
  - $A_1$
  - $A_2$
  - $A_R$

- N experiments
  - Experiment 1
  - Experiment 2
  - Experiment N

- P sensors
  - Sensor 1
  - Sensor 2
  - Prism
  - Sensor P

Matrix formulation

$K_{\text{Signat.}}$ (R VOCs) $\rightarrow$ $C_{\text{Conc.}}$ (R VOCs)

$M_{\text{Masses}}$ (R VOCs) $\rightarrow$ $Y_{\text{Measures}}$ (N exp., P sensors)

$\text{Diagonal}$ Mixture model $Y$
Physico-chemical mixture model
Global model

Problem dimensions

Matrix formulation

\[
\begin{bmatrix}
K \\
\text{Signat.}
\end{bmatrix}
\]

P sensors

R VOCs

A_1, A_2, A_R

P sensors

Sensor 1, Sensor 2, Prism, Sensor P
Physico-chemical mixture model
Global model

Problem dimensions

- **R VOCs**
  - \( A_1 \)
  - \( A_2 \)
  - \( A_R \)

- **N experiments**
  - Experiment 1
  - Experiment 2
  - Experiment N

Matrix formulation

\[
\begin{pmatrix}
K \\
\text{Signat.}
\end{pmatrix}_{R \times P}
\quad
\begin{pmatrix}
C \\
\text{Conc.}
\end{pmatrix}_{N \times P}
\]

- **K**
- **C**
- **N exp.**
Physico-chemical mixture model
Global model

Problem dimensions

Matrix formulation

\[
\begin{align*}
\text{R VOCs} & \quad \text{A}_1 \quad \text{A}_2 \quad \ldots \quad \text{A}_r \\
\text{Problem dimensions} & \quad \text{Matrix formulation}
\end{align*}
\]

\[
\begin{align*}
\begin{bmatrix}
\text{K} \\
\text{Signat.} \\
\text{R VOCs}
\end{bmatrix} & \quad \begin{bmatrix}
\text{C} \\
\text{Conc.} \\
\text{R VOCs}
\end{bmatrix} \\
\text{P sensors} & \quad \text{N exp.}
\end{align*}
\]

\[
\begin{bmatrix}
\text{M} \\
\text{Masses} \\
\text{R VOCs}
\end{bmatrix} \\
\text{R VOCs} \\
\downarrow \\
\text{Diagonal}
\]

\[
\begin{align*}
P & \quad \text{Maho, S. Barthelmé, P. Comon — BSS under the Langmuir model for chemical sensors}
\end{align*}
\]
Physico-chemical mixture model
Global model

Problem dimensions

Matrix formulation

\[
\begin{align*}
K & \quad \text{Signat.} \\
\{ & \quad R \text{ VOCs} \\
\} & \\
C & \quad \text{Conc.} \\
\{ & \quad R \text{ VOCs} \\
\} & \\
M & \quad \text{Masses} \\
\{ & \quad R \text{ VOCs} \\
\} & \\
Y & \quad \text{Measures} \\
\{ & \quad N \text{ exp.} \\
\} & \\
\end{align*}
\]

\[\downarrow \text{Diagonal}\]
### Problem dimensions

<table>
<thead>
<tr>
<th>R VOCs</th>
<th>N experiments</th>
<th>P sensors</th>
</tr>
</thead>
<tbody>
<tr>
<td>A₁, A₂, ... Aᵣ</td>
<td>Experiment 1, Experiment 2, ... Experiment N</td>
<td>Sensor 1, Sensor 2, ... Sensor P</td>
</tr>
</tbody>
</table>

### Matrix formulation

\[
\begin{align*}
\mathbf{K} & \quad \text{Signat.} \\
\mathbf{C} & \quad \text{Conc.} \\
\mathbf{M} & \quad \text{Masses} \\
\mathbf{Y} & \quad \text{Measures}
\end{align*}
\]

\[
\mathbf{Y} = \mathbf{KMC}^t \boxplus (1_P 1_N^t + \mathbf{KC}^t)
\]
Blind Source Separation
Theoretical results

Mixture model

\[ \mathcal{L}(K, C) = Y = KMC^t \boxplus (1_P1_N^t + KC^t) \]

Particular cases

- **Low concentration**: assuming \( KC^t \ll 1 \), then \( Y \approx KMC^t \)
- **Saturation**: assuming high concentrations and/or high affinities, then \( KC^t \gg 1 \) and \( Y \approx \text{highly non-identifiable} \)
Blind Source Separation
Theoretical results

Mixture model

\[ \mathcal{L}(K, C) = Y = KMC^t \boxplus (1_P 1_N^t + KC^t) \]

Particular cases

1. Low concentration: assuming \( KC^t \ll 1 \), then

\[ Y \approx KMC^t \]

\[ \Rightarrow \text{NMF [5]} \]
Blind Source Separation
Theoretical results

Mixture model

\[ \mathcal{L}(K, C) = Y = KM C^t \bigotimes (1_p 1_N^t + KC^t) \]

Particular cases

1. **Low concentration**: assuming \( KC^t \ll 1 \), then
   \( Y \approx KM C^t \)
   \( \rightarrow \) NMF \([5]\]

2. **Saturation**: assuming high concentrations and/or high affinities, then \( KC^t \gg 1 \)
   \( Y \approx KM C^t \bigotimes KC^t \)
   \( \rightarrow \) highly non-identifiable:
   \( Y \approx D_1 KM C^t D_2 \bigotimes D_1 KC^t D_2 \)
Blind Source Separation
Algorithm

Definition of $\Omega$
To avoid the saturation area, we can constrain the product $KC$ by:

$$\min_{K,C} \max_t \left\{ R_{ij}^{pp} KC_{tq} \right\}$$

This constraint is not so easy to implement, so we relax it by:

$$\min_{K,C} \max_t \left\{ K \right\} \max_{q,\omega} \left\{ C \right\}$$

Algorithm
Cost function: $\Upsilon_{p,K,C,q}$

Alternating procedure
0. Initialize $C$
1. Estimate $K$ from the sub-problem: $\arg \min_K \max_t \left\{ R_{ij}^{pp} KC_{tq} \right\}$
2. Estimate $C$ from the sub-problem: $\arg \min_C \max_{q,\omega} \left\{ K \right\}$
3. Repeat from step 1 until convergence
Definition of \( \Omega \)

To avoid the saturation area, we can constrain the product \( KC \) by:

\[
KC \leq \text{max}_{ij} R_{ij} K \quad \text{and} \quad \text{max}_{ij} C \leq \text{max}_{\omega} (K \cdot C)
\]

This constraint is not so easy to implement, so we relax it by:

\[
KC \leq \text{max}_{ij} R_{ij} \quad \text{and} \quad \text{max}_{ij} C \leq \text{max}_{\omega} (K \cdot C)
\]

Algorithm

Cost function: \( \Upsilon \)

Alternating procedure

1. Initialize \( C \)
2. Estimate \( K \) from the sub-problem: \( \arg \min \left\{ K \in \mathbb{R}^+ : \text{max}_{ij} R_{ij} K \right\} \)
3. Estimate \( C \) from the sub-problem: \( \arg \min \left\{ C \in \mathbb{R}^+ : \text{max}_{ij} C \right\} \)
4. Repeat from step 1 until convergence

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Blind Source Separation

Algorithm

\[
\text{Definition of } \Omega
\]

To avoid the saturation area, we can constrain the product \(KC\) by:

\[
\{ KC \} \leq \max_{ij} \{ R_{ij} \} \sup_{pp} \{ KC \} \leq \omega_n
\]

This constraint is not so easy to implement, so we relax it by:

\[
\{ KC \} \leq \max_{ij} \{ K \} \leq \max_{ij} \{ C \} \leq \omega_n
\]

Algorithm

Cost function: \( \Upsilon_{pK,Cq} \)

Alternating procedure

0. Initialize \( C_1 \)

1. Estimate \( K \) from the sub-problem: \( \arg \min K \}

2. Estimate \( C \) from the sub-problem: \( \arg \min C \}

3. Repeat from step 1 until convergence

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Blind Source Separation
Algorithm

Definition of \( \Omega \)

To avoid the saturation area, we can constrain the product \( KC \) by:

\[
KC \leq \max \left\{ R_{ij}^{pp} \right\}
\]

This constraint is not so easy to implement, so we relax it by:

\[
KC \leq \max \left\{ K \right\} \cdot \max \left\{ C \right\}
\]

Algorithm

Cost function: \( \Upsilon_{pK, C_q} \)

Alternating procedure

0. Initialize \( C_1 \)

1. Estimate \( K \) from the sub-problem: \( \arg \min_{K \geq 0} \Upsilon_{pK, C_q} \)

2. Estimate \( C \) from the sub-problem: \( \arg \min_{C \geq 0} \Upsilon_{pK, C_q} \)

3. Repeat from step 1 until convergence
Blind Source Separation

Algorithm

To avoid the saturation area, we can constrain the product $KC$ by:

$$\{ KC \}_{\text{max}} \leq R_{\text{sup}}\{ ij \}_{pp} K \{ \max \}_{\omega} C \{ \max \}_{\omega}$$

This constraint is not so easy to implement, so we relax it by:

$$\{ KC \}_{\text{max}} \leq \{ K \}_{\text{max}} \{ C \}_{\text{max}} \omega$$

**Algorithm**

Cost function: $\Upsilon_k, c_{pq}$

Alternating procedure

0. Initialize $C_1$

1. Estimate $K$ from the sub-problem: $\arg \min K \{ \max \}_{\omega} \{ C \}_{\max} \Upsilon_k, c_{pq}$

2. Estimate $C$ from the sub-problem: $\arg \min C \{ \max \}_{\omega} \{ K \}_{\max} \Upsilon_k, c_{pq}$

3. Repeat from step 1 until convergence
Definition of $\Omega$

To avoid the saturation area, we can constrain the product $KC$ by:

$$\{KC\} \leq \max \{R, \sup_{ij} \rho_{pp} KC, \max_{ij} q \omega K, \max_{ij} C q \omega \}$$

This constraint is not so easy to implement, so we relax it by:

$$\{KC\} \leq \max \{K \{C\} \omega, \max_{ij} \{KC\} \omega \}$$

Algorithm

Cost function: $\Upsilon$.

Alternating procedure

0. Initialize $C$

1. Estimate $K$ from the sub-problem: $\arg \min_K \Upsilon_p K, C q$

2. Estimate $C$ from the sub-problem: $\arg \min_C \Upsilon_p K, C q$

3. Repeat from step 1 until convergence

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Definition of $\Omega$
To avoid the saturation area, we can constrain the product $KC$ by:

$$KC \leq \max\{R_{ij}^{pp} KC_{ij}^{q} \omega^{q-1}\}$$

This constraint is not so easy to implement, so we relax it by:

$$KC \leq \max\{K \leq \max\{C \leq \omega\} \leq \max\{\Upsilon_{p}^{K}, C_{q}^{\Omega}\}$$

Algorithm
Cost function: $\Upsilon^{p} K, C^{q}$

Alternating procedure
0. Initialize $C$
1. Estimate $K$ from the sub-problem: arg min $K \leq 0, \Upsilon^{p} K, C_{q}^{\Omega}$
2. Estimate $C$ from the sub-problem: arg min $C \leq 0, \Upsilon^{p} K, C_{q}^{\Omega}$
3. Repeat from step 1 until convergence

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Definition of $\Omega$

To avoid the saturation area, we can constrain the product $KC$ by:

$$\{KC\}_{t=1}^{\text{max}} \leq \epsilon_{ij}$$

This constraint is not so easy to implement, so we relax it by:

$$\{KC\}_{t=1}^{\text{max}} \leq \epsilon_{ij} \leq \{KC\}_{t=1}^{\text{max}}$$

Algorithm

Cost function: $\gamma_K, \gamma_C$

Alternating procedure

0. Initialize $C$

1. Estimate $K$ from the sub-problem: $\arg\min_K \{KC\}_{t=1}^{\text{max}} \leq \epsilon_{ij} \leq \{KC\}_{t=1}^{\text{max}}$

2. Estimate $C$ from the sub-problem: $\arg\min_C \{KC\}_{t=1}^{\text{max}} \leq \epsilon_{ij} \leq \{KC\}_{t=1}^{\text{max}}$

3. Repeat from step 1 until convergence

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Definition of Ω

To avoid the saturation area, we can constrain the product $KC$ by:

$$\text{max} \{ KC \}$$

This constraint is not so easy to implement, so we relax it by:

$$\text{max} \{ K \} \text{max} \{ C \}$$

Algorithm

Cost function: $\Upsilon_p K, C$}

$\Upsilon_p K, C$

Alternating procedure

0 Initialize $C$

1 Estimate $K$ from the sub-problem: arg min $K \in \mathbb{R}$, $C \in \mathbb{R}$

2 Estimate $C$ from the sub-problem: arg min $C \in \mathbb{R}$, $K \in \mathbb{R}$

Repeat from step 1 until convergence
Definition of $\Omega$

To avoid the saturation area, we can constrain the product $KC$ by:

$$\|KC^t\|_{\text{max}} = R \sup_{ij} ((KC^t)_{ij}) \leq \omega$$

This constraint is not so easy to implement, so we relax it by:

$$\|KC^t\|_{\text{max}} \leq \|K\|_{\text{max}} \|C^t\|_{\text{max}} \leq \omega$$
Definition of $\Omega$

To avoid the saturation area, we can constrain the product $KC$ by:

$$\|KC^t\|_{\text{max}} = R \sup_{ij} (KC^t)_{ij} \leq \omega$$

This constraint is not so easy to implement, so we relax it by:

$$\|KC^t\|_{\text{max}} \leq \|K\|_{\text{max}} \|C^t\|_{\text{max}} \leq \omega$$

Algorithm

Cost function:

$$\Upsilon(K, C) = \|Y - \mathcal{L}(K, C)\|_F$$

Alternating procedure

0 Initialize $C$

1 Estimate $K$ from the sub-problem:

$$\arg\min_{K \geq 0, \|K\|_{\text{max}} \leq \|C^\omega\|_{\text{max}}} \Upsilon(K, C)$$

2 Estimate $C$ from the sub-problem:

$$\arg\min_{C \geq 0, \|C\|_{\text{max}} \leq \|K^\omega\|_{\text{max}}} \Upsilon(K, C)$$

3 Repeat from step 1 until convergence
Simulation results

Simulation settings

<table>
<thead>
<tr>
<th>R VOCS</th>
<th>5/10/15</th>
</tr>
</thead>
<tbody>
<tr>
<td>N experiments</td>
<td>100</td>
</tr>
<tr>
<td>P sensors</td>
<td>100</td>
</tr>
</tbody>
</table>

Data are simulated with an additional Gaussian noise:

\[ Y = \mathcal{L}(K, C) + \epsilon \text{ with } \epsilon_{ij} \sim \mathcal{N}(0, \sigma_n) \]

The noise is progressively intensified by decreasing the following Signal to Noise Ratio (SNR):

\[ \text{SNR} = 20 \log\left(\frac{\sigma_s}{\sigma_n}\right) \text{ with } \sigma_s = \sqrt{\frac{\|Y\|^2}{PN}} \]
Conclusion

We have

① formulated a non-linear mixture model for a type of chemical sensors used in an electronic nose.
② proposed an algorithm in order to estimate blindly the individual outputs and the concentrations.
③ assessed the performance of the algorithm in the presence of noise.

Further work will

① include experiments with real data.
② relax the assumption that we know the masses.
③ exploit time information.
References


Acknowledgements

The authors would like to acknowledge Cyril Herrier and Thierry Livache from the start-up Aryballe Technologies and Arnaud Buhot from the CEA Inac for their highly valuable help in the understanding of the proposed model.
Thank you ! Questions ?