Analyse comparative d'algorithmes de restauration en architecture dépliée pour des signaux chromatographiques parcimonieux

(+bonus: MLNIR, near-infrared spectra/densities open data)

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Of people and projects

- People:
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- Projects:
  - EU H2020-MSCA-ETN TraDE-OPT-861137;
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  - ERC Consolidator Grant SLING 819789;
  - EU H2020-MSCA-RISE NoMADS-777826.

Outline

Additional information

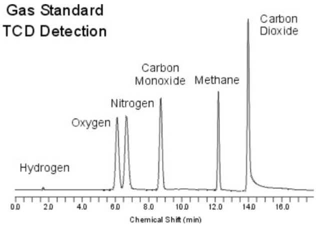
- Unrolling and un/self/*/supervised learning for inverse problems (GdR IASIS, May 13th, 2025)
  - gdr-iasis.cnrs.fr/reunions/unrolling-and-un-self-supervised-learning-for-inverse-problems/
- More on unrolling/unfolding?
  - Monday 25th, 2025, 17:00 - 18:40 : Thème 3 - Poster 1 : Localisation et séparation de sources
    ID1621 : Mises à jour multiplicatives dépliées pour la Factorisation en Matrices Non-négatives appliquée au démélange spectral.
    Christophe Kervazo, Jérémy Cohen

Ìn kùrzer Zitt

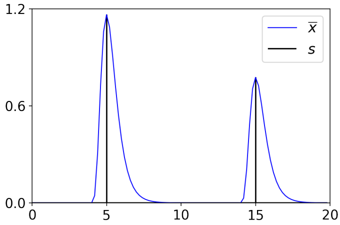
Ìn kùrzer Zitt: analytical chemistry and chromatography

Analytical chemistry: Study of chemical composition, characteristics of compounds, properties...

Chromatography: separation of a mixture into components based on mobile vs stationary phase (retention time).



Chromatographics peaks: ($\pm$ sparse) toy peak model: quality, quantity.

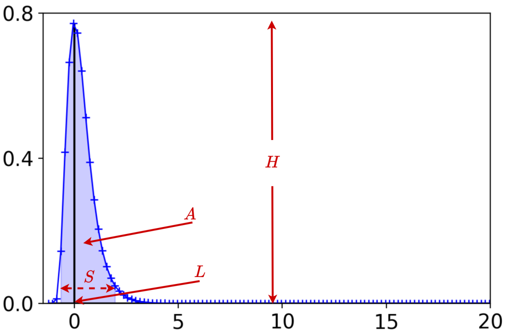


$\longrightarrow$ Indirect **sparse** information + model + uncertainty: inverse problem

Ìn kùrzer Zitt: comprehensive database and HALmetrics

Sparsity hypothesis:

- MSE, SNR, or Truncated SNR may not suffice;
- HALmetrics: peak-based Height-Area-Location evaluation.



Background

Notations and challenges

Inverse problem in analytical chemistry

$$\mathbf{z} = \mathbf{H}\overline{\mathbf{x}} + \mathbf{e},$$

- $\mathbf{z} \in \mathbb{R}^M$: observed acquisition,
- $\overline{\mathbf{x}} \in \mathbb{R}^N$: original sparse positive-valued signal,
- $\mathbf{H} \in \mathbb{R}^{M \times N}$: measurement degradation, typically convolution with Gaussian, Voigt, **Fraser-Suzuki**... kernel shapes,
- $\mathbf{e}$: corrupting noise, typically additive Gaussian iid.

⟶ **To retrieve an estimate $\widehat{\mathbf{x}} \in \mathbb{R}^N$ of $\overline{\mathbf{x}} \in \mathbb{R}^N$ knowing $\mathbf{H}$ and $\mathbf{z}$.**

Challenges:

- Heterogenous signals
- High dimensionality
- Large databases

State-of-the-art methods

Goal: Find an estimate $\widehat{\mathbf{x}}$ of $\overline{\mathbf{x}}$ from knowledge of $\mathbf{z}$ and $\mathbf{H}$.

I. Model-based

$$\widehat{\mathbf{x}} \in \underset{\mathbf{x}\in\mathbb{R}^N}{\operatorname{argmin}} \; \underbrace{J(\mathbf{Hx},\mathbf{z})}_{\text{Data fidelity}} + \underbrace{\lambda\Psi(\mathbf{x})}_{\text{Regularization}}$$

$\Rightarrow$ Iterative optimization algorithm

II. Learning-based

$$\widehat{\mathbf{x}} = \underbrace{h(\mathbf{z},\widehat{\Theta})}_{\text{Mapping}}$$

$\Rightarrow$ Backpropagation

State-of-the-art methods

Model-based	Learning-based
✓ Theoretical guarantees	✓ Good accuracy
✓ Robustness	✓ Easy deployment
✓ Explainability	✓ Fast (at test phase)
✗ High computational cost	✗ Lack of robustness
✗ Tedious parameter tuning	✗ Black-Box: Not explainable

⟶ Get the best out of the two approaches?

Unrolling/Unfolding methods [Bertocchi et al., 2020.]

❶ Initial Algorithm

1: Init: Choose Θ, $\mathbf{x}_0 \in \mathbb{R}^N$.
2: **for** $k = 0, 1, \ldots$ **do**
3: $\quad \mathbf{x}_{k+1} = \mathcal{I}_k^{(\Theta)}(\mathbf{x}_k, \mathbf{z})$,
4: **end for**
5: Return $\widehat{\mathbf{x}}$.

❷ Reinterpretation

- Truncate the number of iterations to a fixed value of layers K:

$$\textbf{Iteration } \mathcal{I}_k^{(\Theta)}(\cdot, \mathbf{z}) : \mathbb{R}^N \to \mathbb{R}^N \iff \textbf{Layer } \mathcal{L}_k^{(\Theta_k)}(\cdot, \mathbf{z}) : \mathbb{R}^N \to \mathbb{R}^N.$$

❸ Learning and inferring

- Training: Minimize task-oriented loss ℓ between groudtruths and estimates of unrolled architecture with respect to $(\Theta_k)_{\{0 \leq k \leq K-1\}}$.
- Test: $\widehat{\mathbf{x}} = \mathcal{L}_{K-1}^{(\widehat{\Theta}_{K-1})}(\cdot, \mathbf{z}) \circ \cdots \circ \mathcal{L}_0^{(\widehat{\Theta}_0)}(\cdot, \mathbf{z})(\mathbf{x}_0)$.

Learning Strategy

1. Training Set: $\{(\overline{\mathbf{x}}^{(i)}, \mathbf{z}^{(i)}),\ i \in \{1, \ldots, S\}\}$
2. Feedforward model:

$$f_\Theta(\mathbf{x}_0^{(i)}; \mathbf{z}^{(i)}) = \mathcal{L}_{K-1}^{\mathbf{z}^{(i)}} \circ \cdots \circ \mathcal{L}_k^{\mathbf{z}^{(i)}} \circ \cdots \circ \mathcal{L}_0^{\mathbf{z}^{(i)}}(\mathbf{x}_0^{(i)})$$

3. Backpropagation

$$\widehat{\Theta} = \underset{\Theta \in \mathbb{R}^{K(2M+L)}}{\operatorname{argmin}} \quad \frac{1}{S} \sum_{i=1}^{S} \ell(f_\Theta(\mathbf{x}_0^{(i)}; \mathbf{z}^{(i)}), \overline{\mathbf{x}}^{(i)})$$

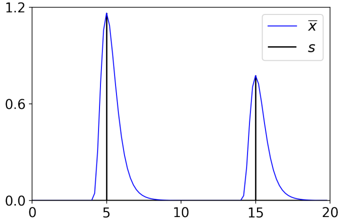
4. Inference/test: $\widehat{\mathbf{x}} = f_{\widehat{\Theta}}(\mathbf{x}_0; \mathbf{z})$

Problem

Motivation

- **Inverse problem**

$$\mathbf{z} = \mathbf{H}(\underbrace{\pi * \mathbf{s}}_{\bar{\mathbf{x}}}) + \mathbf{e}$$

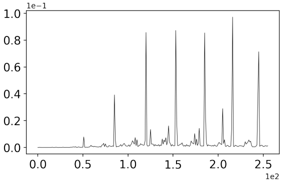


- **Goal:**
  U-HQ, **U-ISTA** and **U-PD** on chromatographic datasets (parametric and real) using chemically-driven evaluation metrics (HALmetrics) to recover an estimate $\widehat{\mathbf{x}}$ of $\bar{\mathbf{x}}$.

Data

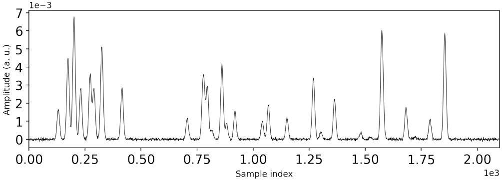
Parametric model inspired from real data

- **Actual chromatogram shape**



- **Peak modelling: Fraser-Suzuki chromatogram simulation**

$$\left(\forall x > m - \frac{\sigma_f}{a}\right): \quad \pi(x) \propto \exp\left(-\frac{1}{2a^2}\log\left(1 + a\frac{(x-m)}{\sigma_f}\right)^2\right).$$



Parametric databases: parameters

- For spiky signal $\mathbf{s}$:
  - number of samples N;
  - number of spikes or sparsity level P; expressed relatively as P/N;
  - peak separation limit $d_{\mathrm{min}} \in \{1, \ldots, N\}$;
  - spike intensities: $|\mathcal{N}(0,1)|$ (absolute value of a standard normal distribution);
- For peak kernel π:
  - peak width $\sigma_f > 0$;
  - asymmetric tailing coefficient $a > 0$ ($a \longrightarrow 0$: Gaussian peak);
- For external disturbance sources:
  - additive "noise": zero-mean Gaussian, standard deviation $\sigma_{\mathbf{e}} > 0$;
  - potential instrument response blur kernel with width $\sigma_{\mathbf{G}} > 0$.

Parametric databases: summary

- **Parameters**
  - Signal **s**: N, P, d_{min};
  - Peak kernel π: (σ_f), a;
  - External disturbances: $\sigma_{\mathbf{e}}$, $(\sigma_{\mathbf{H}})$.
- **Dataset summary**

Parameter\Dataset	D0	D1	D2	D3	D4	D5	D6
P/N	1.5%	3%	4.5%	1.5%	1.5%	3%	3%
d_{min}	5	3	1	5	5	3	3
(σ_f)	0.5	0.5	0.5	0.5	0.5	0.5	0.5
a	0.2	0.2	0.2	0.4	0.6	0.2	0.2
$\sigma_{\mathbf{e}}$	0.02	0.02	0.02	0.02	0.02	0.04	0.06
$(\sigma_{\mathbf{H}})$	1	1	1	1	1	1	1

$\longrightarrow$ Several datasets with different sparsity, peak shape and noise level.

Parametric databases: design of experiments (DoE)

- **Parameters**
  - Signal **s**: N, P, d_{min}.
  - Peak kernel π: σ_f, a.
  - External disturbances: $\sigma_{\mathbf{e}}$ ($\sigma_{\mathbf{H}}$).
- **Dataset DoE difficulty**

Variation\Difficulty	Low	Mid	High
Sparsity ($P/N, d_{\min}$)	D0 ($1.5\%, 5$)	D1 ($3\%, 3$)	D2 ($4.5\%, 1$)
Asymmetry (a)	D0 (0.2)	D3 (0.4)	D4 (0.6)
Noise ($\sigma_{\mathbf{e}}$)	D1 (0.02)	D5 (0.04)	D6 (0.06)

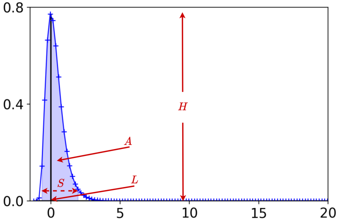
Table 1: DoE. Parametric variations on sparsity, asymmetry and noise.

Evaluation: classical signal metrics

$$\mathrm{MSE}(\mathbf{p}, \widehat{\mathbf{p}}) = \frac{1}{n} \|\mathbf{p} - \widehat{\mathbf{p}}\|^2,$$

$$\mathrm{SNR}(\mathbf{p}, \widehat{\mathbf{p}}) = 20 \log_{10} \left(\frac{\|\mathbf{p}\|}{\|\mathbf{p} - \widehat{\mathbf{p}}\|} \right),$$

$$\mathrm{TSNR}(\mathbf{p}, \widehat{\mathbf{p}}) = 20 \log_{10} \left(\frac{\sum_i |\mathbf{p}^{(i)}|^2}{\sum_i |\mathbf{p}^{(i)} - \widehat{\mathbf{p}^{(i)}}|^2} \right), \quad \forall i \in \bigcup_{1 \le j \le P} \mathcal{S}_j \,.$$



Evaluation: peak HALmetric quantities

- Ground-truth quantities

$$\overline{L_j} = \arg \max_{i \in \{1,\dots,N\}} \left((s \odot \delta_{S_j}) * \pi \right)_{1 \le i \le N},$$

$$\overline{H_j} = p_{\overline{L_j}},$$

$$\mathcal{S}_j = \{ i \in \{1, \dots, N\} \quad \text{s.t.} \quad (s \odot \delta_{S_j}) * \pi \ge \vartheta \overline{H_j} \} = [aj \cdot \cdot bj],$$

$$\overline{A_j} = \sum_{i \in \mathcal{S}_j \setminus b_j} (p_i + p_{i+1})/2.$$

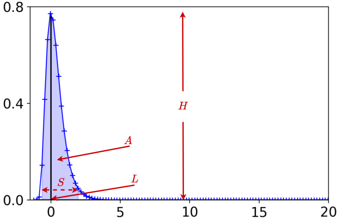
- Estimates using oracle $\mathcal{S}_j$

$$\widehat{L_j} = \arg \max_{i \in \mathcal{S}_j} \hat{p}_i,$$

$$\widehat{H_j} = \hat{p}_{\widehat{L_j}},$$

$$\widehat{A_j} = \sum_{i \in \mathcal{S}_j \setminus b_j} (\hat{p}_i + \hat{p}_{i+1})/2.$$

Evaluation: peak HALmetrics

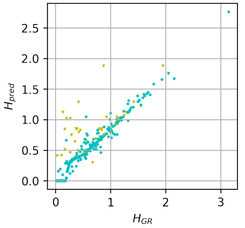


- Objective (relative) HALmetrics:
  Given $\widehat{\mathbf{H}}$, $\overline{\mathbf{H}}$, we define:

$$\text{NMAE}(\overline{\mathbf{H}}, \widehat{\mathbf{H}}) = \frac{\sum_{j=1}^{P} |\overline{H_j} - \widehat{H_j}|}{\sum_{j=1}^{P} |\overline{H_j}|}.$$

Sim.: $\text{NMAE}(\overline{\mathbf{L}}, \widehat{\mathbf{L}}) / \text{NMAE}(\overline{\mathbf{A}}, \widehat{\mathbf{A}})$.

- Subjective HALmetrics:
  Scatter plots.

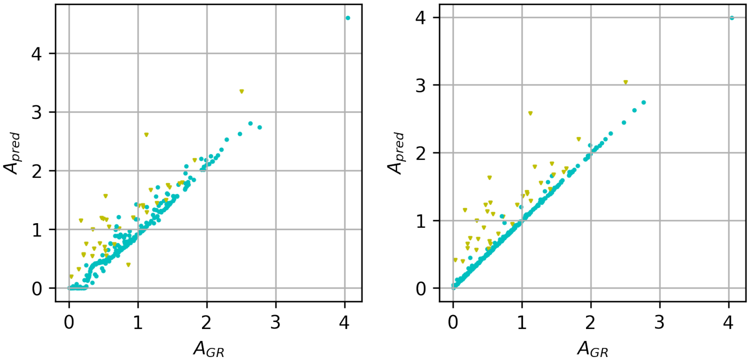


Results

Results: classical and objective HALmetrics

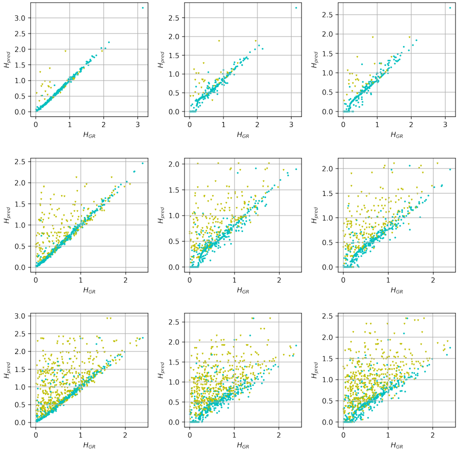
		MSE	SNR	TSNR	NMAE($\bar{\mathbf{H}}, \hat{\mathbf{H}}$)	NMAE($\bar{\mathbf{A}}, \hat{\mathbf{A}}$)	NMAE($\bar{\mathbf{L}}, \hat{\mathbf{L}}$)
D0	**U-HQ**	$\mathbf{4.7 \times 10^{-4}}$ $\mathbf{(1.2 \times 10^{-4})}$	**19.6 (0.9)**	**20.0 (1.0)**	**0.1 (0.0)**	**0.012 (0.003)**	$\mathbf{3.1 \times 10^{-6}}$ $\mathbf{(1.5 \times 10^{-6})}$
	U-PD	3.7×10^{-3} (1.6×10^{-3})	10.9 (1.3)	10.9 (1.3)	0.2 (0.0)	0.107 (0.025)	8.0×10^{-6} (1.8×10^{-6})
	U-ISTA	3.0×10^{-3} (1.0×10^{-3})	11.7 (1.0)	11.7 (1.0)	0.2 (0.0)	0.110 (0.020)	8.0×10^{-6} (1.7×10^{-6})
D1	**U-HQ**	$\mathbf{1.1 \times 10^{-3}}$ $\mathbf{(2.3 \times 10^{-4})}$	**19.4 (0.7)**	**19.6 (0.7)**	**0.2 (0.1)**	**0.014 (0.002)**	$\mathbf{3.8 \times 10^{-6}}$ $\mathbf{(9.5 \times 10^{-7})}$
	U-PD	10.0×10^{-3} (2.5×10^{-3})	9.7 (0.7)	9.7 (0.7)	0.4 (0.0)	0.107 (0.016)	6.3×10^{-6} (8.7×10^{-7})
	U-ISTA	8.0×10^{-3} (2.1×10^{-3})	10.6 (0.7)	10.6 (0.7)	0.3 (0.0)	0.109 (0.019)	6.1×10^{-6} (8.6×10^{-7})
D2	**U-HQ**	$\mathbf{1.7 \times 10^{-3}}$ $\mathbf{(3.4 \times 10^{-4})}$	**19.6 (0.8)**	**19.7 (0.8)**	**0.4 (0.1)**	**0.015 (0.002)**	$\mathbf{5.2 \times 10^{-6}}$ $\mathbf{(9.5 \times 10^{-7})}$
	U-PD	1.7×10^{-2} (3.3×10^{-3})	9.5 (0.5)	9.5 (0.5)	0.5 (0.1)	0.101 (0.012)	5.8×10^{-6} (7.2×10^{-7})
	U-ISTA	1.4×10^{-2} (2.6×10^{-3})	10.4 (0.6)	10.4 (0.6)	0.5 (0.1)	0.102 (0.012)	5.8×10^{-6} (7.5×10^{-7})
D3	**U-HQ**	$\mathbf{8.7 \times 10^{-4}}$ $\mathbf{(2.1 \times 10^{-4})}$	**16.9 (0.7)**	**17.3 (0.8)**	**0.1 (0.1)**	**0.015 (0.002)**	$\mathbf{3.7 \times 10^{-6}}$ $\mathbf{(1.7 \times 10^{-6})}$
	U-PD	4.4×10^{-3} (1.4×10^{-3})	10.0 (0.9)	10.1 (0.9)	0.3 (0.0)	0.107 (0.020)	9.0×10^{-6} (1.7×10^{-6})
	U-ISTA	3.7×10^{-3} (1.1×10^{-3})	10.7 (0.7)	10.7 (0.7)	0.2 (0.0)	0.104 (0.027)	8.7×10^{-6} (1.7×10^{-6})
D4	**U-HQ**	$\mathbf{1.4 \times 10^{-3}}$ $\mathbf{(2.8 \times 10^{-4})}$	**14.9 (0.6)**	**15.1 (0.6)**	**0.1 (0.1)**	**0.012 (0.002)**	$\mathbf{4.5 \times 10^{-6}}$ $\mathbf{(1.9 \times 10^{-6})}$
	U-PD	5.0×10^{-3} (1.4×10^{-3})	9.3 (0.5)	9.3 (0.5)	0.3 (0.0)	0.1 (0.0)	9.6×10^{-6} (1.6×10^{-6})
	U-ISTA	4.5×10^{-3} (1.2×10^{-3})	9.8 (0.6)	9.8 (0.6)	0.3 (0.0)	0.083 (0.011)	9.3×10^{-6} (1.6×10^{-6})
D5	**U-HQ**	$\mathbf{1.5 \times 10^{-3}}$ $\mathbf{(3.3 \times 10^{-4})}$	**17.8 (0.7)**	**18.0 (0.8)**	**0.2 (0.1)**	**0.018 (0.003)**	$\mathbf{4.2 \times 10^{-6}}$ $\mathbf{(1.0 \times 10^{-6})}$
	U-PD	10.0×10^{-3} (2.3×10^{-3})	9.6 (0.7)	9.6 (0.7)	0.4 (0.0)	0.108 (0.015)	6.5×10^{-6} (9.1×10^{-7})
	U-ISTA	7.9×10^{-3} (1.8×10^{-3})	10.6 (0.6)	10.6 (0.6)	0.4 (0.0)	0.106 (0.014)	6.3×10^{-6} (9.1×10^{-7})
D6	**U-HQ**	$\mathbf{1.9 \times 10^{-3}}$ $\mathbf{(4.0 \times 10^{-4})}$	**16.8 (0.8)**	**17.1 (0.8)**	**0.2 (0.1)**	**0.021 (0.003)**	$\mathbf{4.2 \times 10^{-6}}$ $\mathbf{(9.8 \times 10^{-7})}$
	U-PD	9.9×10^{-3} (2.3×10^{-3})	9.7 (0.7)	9.7 (0.7)	0.4 (0.0)	0.108 (0.016)	6.3×10^{-6} (8.7×10^{-7})
	U-ISTA	7.9×10^{-3} (1.7×10^{-3})	10.6 (0.6)	10.7 (0.6)	0.3 (0.0)	0.107 (0.014)	6.1×10^{-6} (8.6×10^{-7})

Results: subjective HALmetrics interpretation



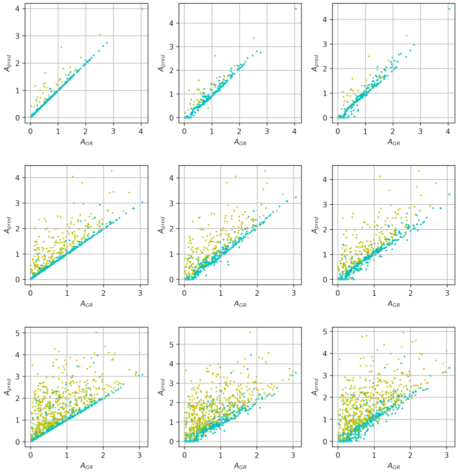
Subjective HALmetrics. Scatter plots of predicted area under curve $\hat{\mathbf{A}}$ with respect to ground truth $\overline{\mathbf{A}}$, for some data set with Method 1 (left) and Method 2 (right).

Results: subjective HALmetrics evaluation, height (H)



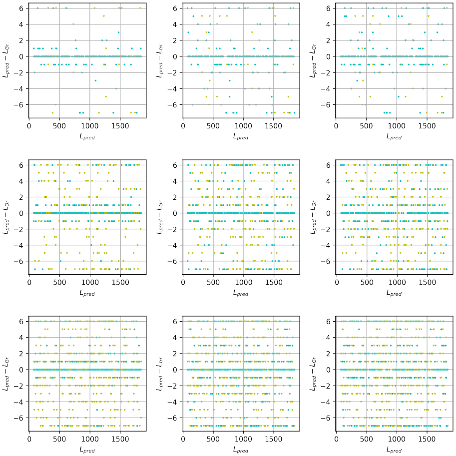
Scatter plots: $(\overline{\mathbf{H}}, \hat{\mathbf{H}})$. Columns: **U-HQ**, **U-ISTA**, **U-PD**. Rows: sparsity ($D0$, $D1$, $D2$).

Results: subjective HALmetrics evaluation, area (A)



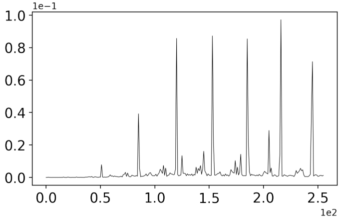
Scatter plots: $(\overline{\mathbf{A}}, \hat{\mathbf{A}})$. Columns: **U-HQ**, **U-ISTA**, **U-PD**. Rows: sparsity ($D0$, $D1$, $D2$).

Results: subjective HALmetrics evaluation, location (L)

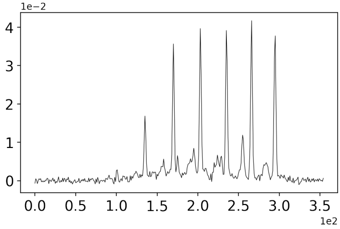


Scatter plots: $(\bar{\mathbf{L}}, \hat{\mathbf{L}})$. Columns: **U-HQ**, **U-ISTA**, **U-PD**. Rows: sparsity ($D0$, $D1$, $D2$).

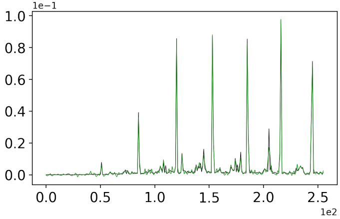
Reconstruction on Real Data U-HQ vs U-ISTA



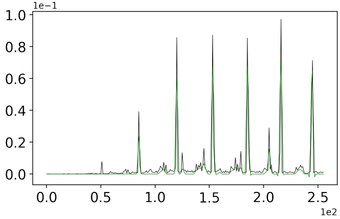
Original



Degraded



U-HQ (17.99 dB)

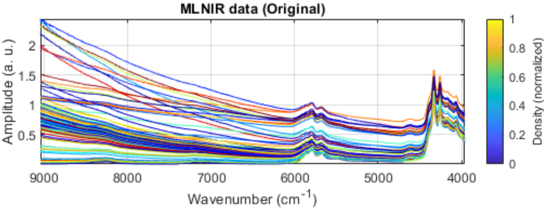


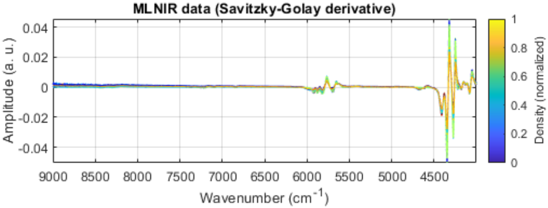
U-ISTA (4.90 dB)

Conclusion

- Code: github.com/GHARBIMouna/Unrolled-Half-Quadratic/.
- Benchmark of a novel neural network: unrolling a majorization-minimization half-quadratic algorithm named **U-HQ**.
- Automatic learning of regularization parameters and stepsize.
- Benchmarking of three well-established methods for sparse signal recovery and their unrolled counterparts through chemically-driven metrics to perform a comprehensive study of unrolling.

MLNIRdata: an open data bonus





MLNIRdata: open data with 208 NIR spectra and densities.
https://doi.org/10.5281/zenodo.16781222

Related publications

Journal articles

- M. Gharbi, E. Chouzenoux, J.-C. Pesquet. An unrolled half-quadratic approach for sparse signal recovery in spectroscopy. Signal Processing, 2024, no. 218, pp. 109369.

Work in progress

- M. Gharbi, S. Villa, E. Chouzenoux, J.-C. Pesquet, L. Duval. A comprehensive study of unrolling through chromatographic data. To be submitted, Sep. 2025.

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H. H. Bauschke and P. L. Combettes. *Convex Analysis and Monotone Operator Theory in Hilbert Spaces*. Springer Publishing Company, Incorporated, 2nd edition, 2017.

C. Bertocchi, E. Chouzenoux, M. Corbineau, J.-C. Pesquet, and M. Prato. Deep Unfolding of a Proximal Interior Point Method for Image Restoration. *Inverse Problems*, 36(3):034005, 2020.

P. Combettes and J.-C. Pesquet. Lipschitz certificates for layered network structures driven by averaged activation operators. *SIAM Journal on Mathematics of Data Science*, 2:529–557, Jun 2020.