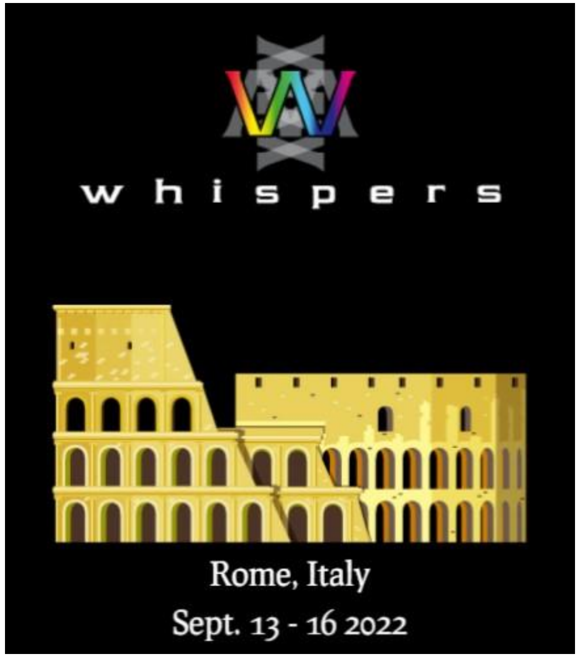
# Unveiling “The Scream” by Edvard Munch: Iterative Fuzzy C-means Analysis Of Macro-XRF Mapping




M.L. Cardinali, F. Albertin, L. Cartechini, I. C. A. Sandu, E. Storevik Tveit, A. Romani, C. Grazia, R. P. De Freitas, V. Bruni, D. Vitulano, F. Rosi

SAPIENZA UNIVERSITÀ DI ROMA
SCITEC ISTITUTO DI SCIENZE E TECNOLOGIE CHIMICHE GIULIO NATTA
A.D. 1308 unipg DIPARTIMENTO DI CHIMICA, BIOLOGIA E BIOTECNOLOGIE
INSTITUTO FEDERAL Rio de Janeiro Campus Paracambi

# Outline

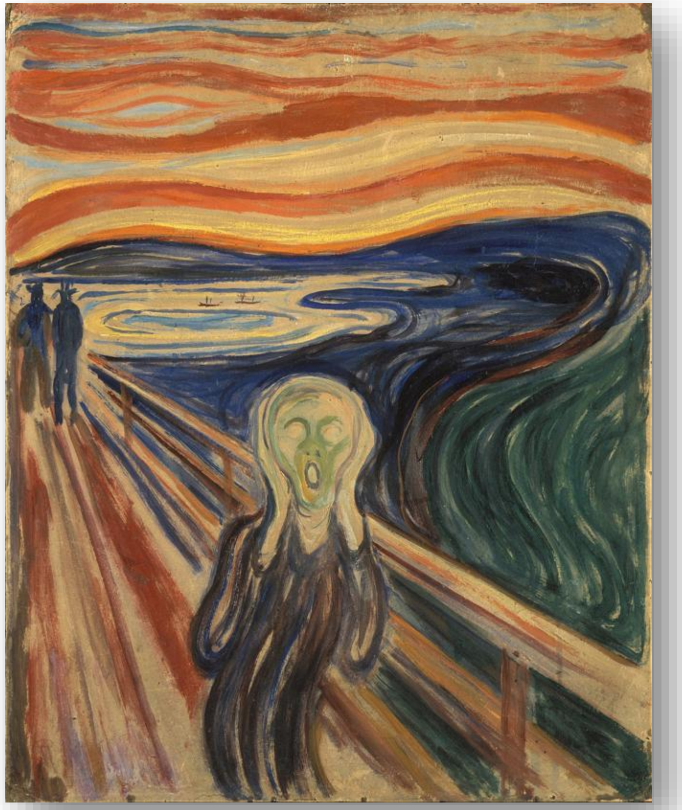
1. Data description and preliminary analysis
2. Algorithm:

   **FCM**

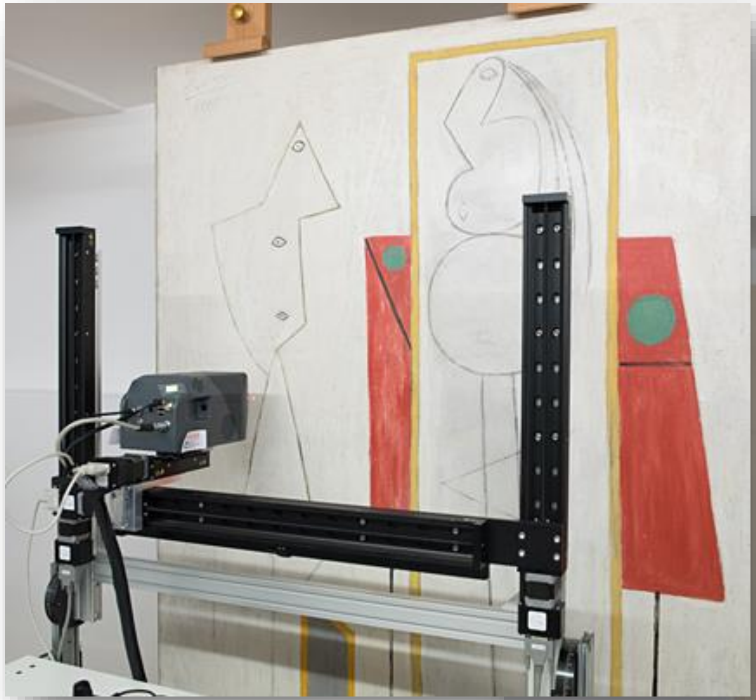
3. Procedure and settings:

   **Two-stage FCM-based analysis**

4. Results
5. Conclusion and future perspectives



# MA-XRF investigations



## MA-XRF:

Widely used non-invasive and non-destructive technique available in situ

## IN CULTURAL HERITAGE

- Elemental analysis and mapping of works of art materials
- Information about dates, artist techniques, restoration interventions, clues of fakes

# MA-XRF investigations


## MA-XRF Scanner:

### CRONO by XGLaB (Bruker Nano Analytics)

- **Source**: X-ray tube with Rh anode, max 50KV 200 μA, 1 or 2mm pinhole collimator
- **Detector**: silicon drift detector Energy resolution 130eV at MnKa, 4096 channels
- **Scanning system**: Scan Area 60x45cm, Max Speed: 30 mm/s; Z range: 7.5 cm

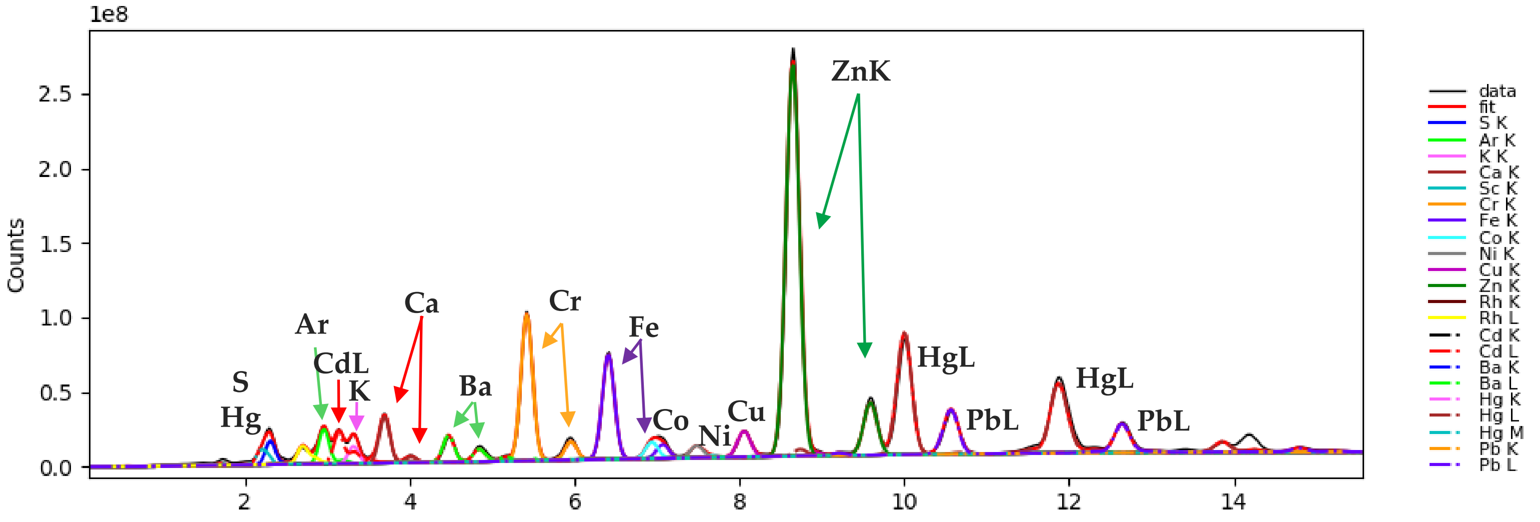
### MUNCH acquisition set-up:

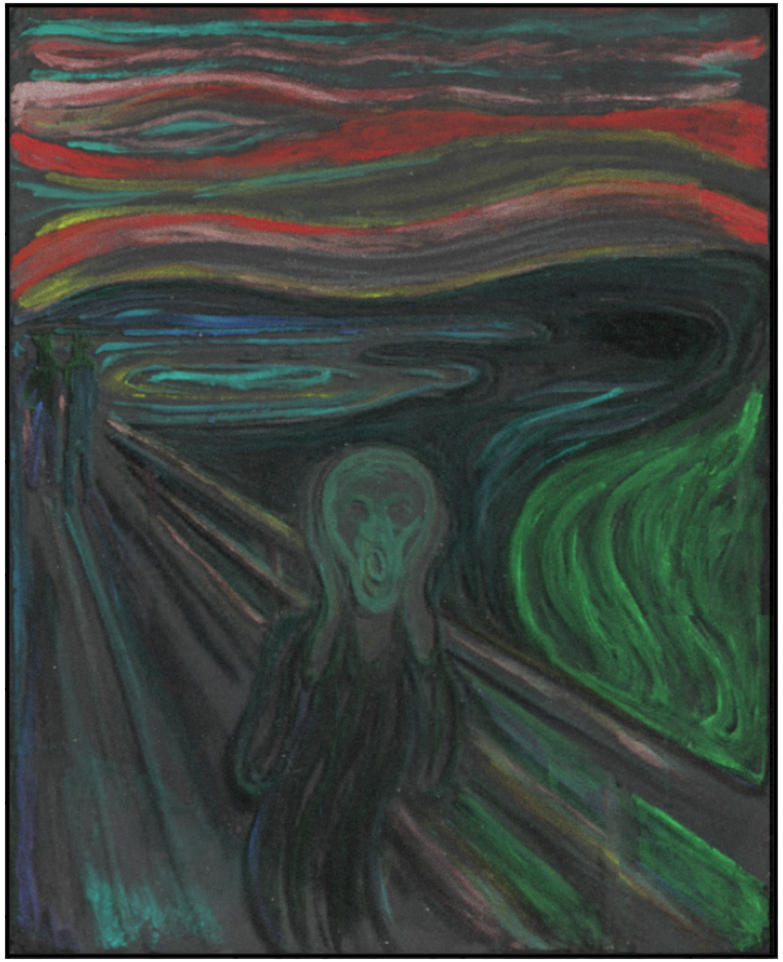
- Four data-taking sessions to cover the entire painting
- **Source**: 50KV, 200 μ A, 1mm collimator
- **Scanning parameters**: linear speed 20 mm/s, 40ms per spectrum

### Complete Map

835 x 675 pixels/spectra binned from 4096 to 2048 channels to improve the signal-to-noise ratio

# Preliminary analysis of the MA-XRF datacube




**Softwares:** PyMCA [1], DataMuncher [2]

*[1] M. Cotte et al., Watching Kinetic Studies as Chemical Maps Using Open-Source Software, Anal. Chem. 88 (2016) 6154–6160;*

*[2] M. Alfeld, K. Janssens, Strategies for processing mega-pixel x-ray fluorescence hyperspectral data: A case study on a version of Caravaggio's painting, Supper at Emmaus. J. Anal. At. Spectrom 30, 777–789 (2015);*

# Preliminary analysis of the MA-XRF datacube

**LIMITS:**

Not able to provide sufficient information about the mixtures of the identified pigments and the manifold paint layering

# Preliminary analysis of the MA-XRF datacube

**LIMITS:**

Not able to provide sufficient information about the mixtures of the identified pigments and the manifold paint layering

**Iterative and nonlinear process:**

**blind FCM + supervised analysis**

# Fuzzy C-Means (FCM) algorithm

- Unsupervised clustering technique
- Points can belong to multiple clusters with varying degrees of membership [3]

*[3] J.C. Bezdek, "Pattern Recognition with Fuzzy Objective Function Algorithms", Plenum Press, New York, 1981.*

# Fuzzy C-Means (FCM) algorithm

- Unsupervised clustering technique
- Points can belong to multiple clusters with varying degrees of membership [3]

**IN THE CONTEXT OF SPECTRAL IMAGES:**

- identification of structures in the data
- segmentation of the image into regions with similar spectral properties

*[3] J.C. Bezdek, "Pattern Recognition with Fuzzy Objective Function Algorithms", Plenum Press, New York, 1981.*

# Fuzzy C-Means (FCM) algorithm

Minimization of the **objective function**:

$$J_m = \sum_{i=1}^{D} \sum_{j=1}^{N} \mu_{ij}^{m} \left\| x_i - c_j \right\|^2$$

where:

- $D$ **number of data points**, $N$ **number of clusters**
- $x_i$ **$i$th data point**, $c_j$ **center of the $j$th cluster**
- $m$ **fuzzy partition matrix exponent**, $\mu_{ij}$ **degree of membership** of $x_i$ in the $j$th cluster

The algorithm stops when: **maximum number of iterations** $max_{iter}$ reached
**OR** the variation of the objective function less than **predefined threshold** $\Delta_J$

# Fuzzy C-Means (FCM) algorithm

**INPUT:** *number of clusters* $N$*, fuzzy exponent* $m$*, maximum number of iterations* $max_{iter}$*, threshold* $\Delta_J$

1. *Initialization of the membership values,* $\mu_{ij}$.
2. ***While** the maximum number of iteration* $max_{iter}$ *is not reached*
   *and the variation of the objective function* $J_m$ *is not less than the threshold* $\Delta_J$*:*

   *2.1. Computation of cluster centers:*

$$c_j = \frac{\sum_{i=1}^{D} {\mu_{ij}}^m x_i}{\sum_{i=1}^{D} {\mu_{ij}}^m} ;$$

   *2.2. Updating of* $\mu_{ij}$*:*

$$\mu_{ij} = \frac{1}{\sum_{k=1}^{N} \left( \frac{\|x_i - c_j\|}{\|x_i - c_k\|} \right)^{\frac{2}{m-1}}} ;$$

   *2.3. Evaluation of the objective function* $J_m$ *and its variation.*

**OUTPUT:** *Soft partition* $P$

Conversion of the soft partition to a hard partition according to a **maximum membership rule.**

# Two-stage FCM-based analysis

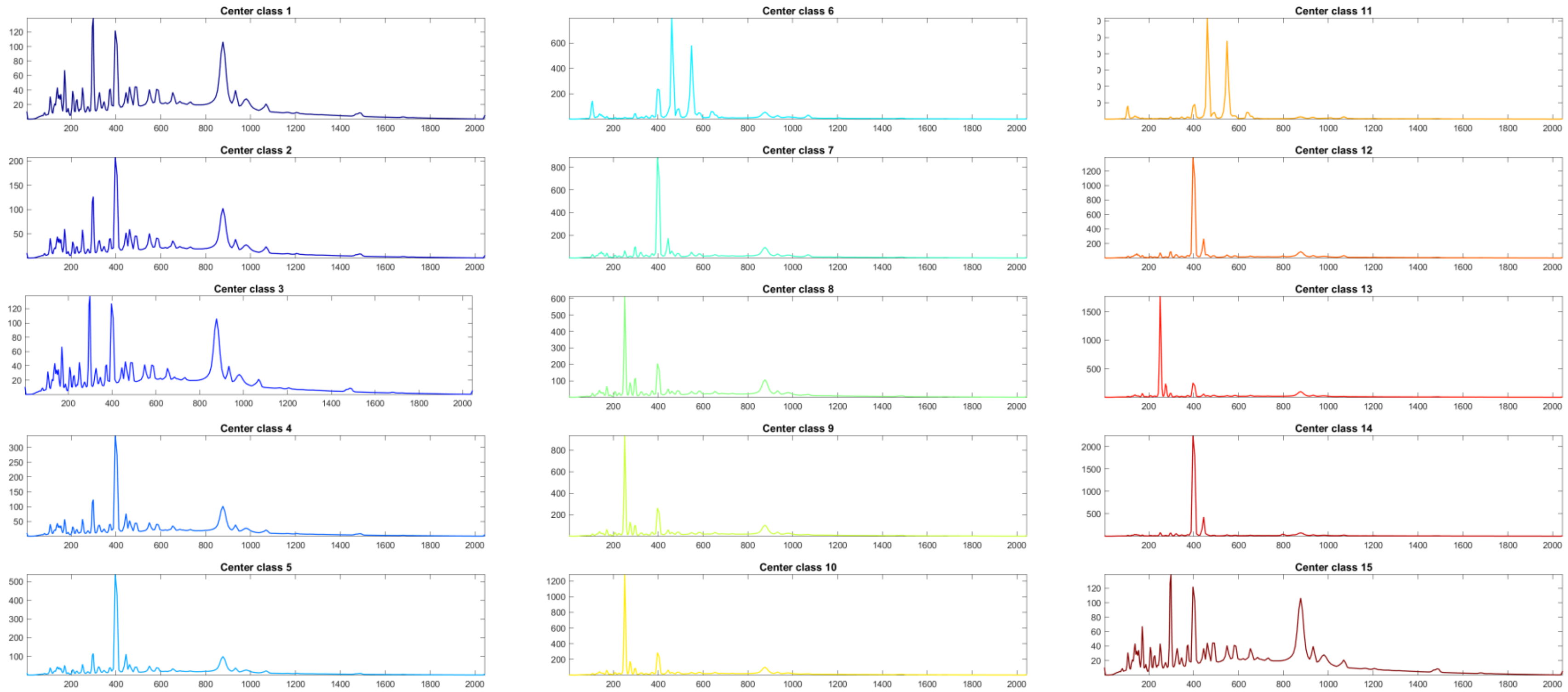
**FIRST STAGE:**

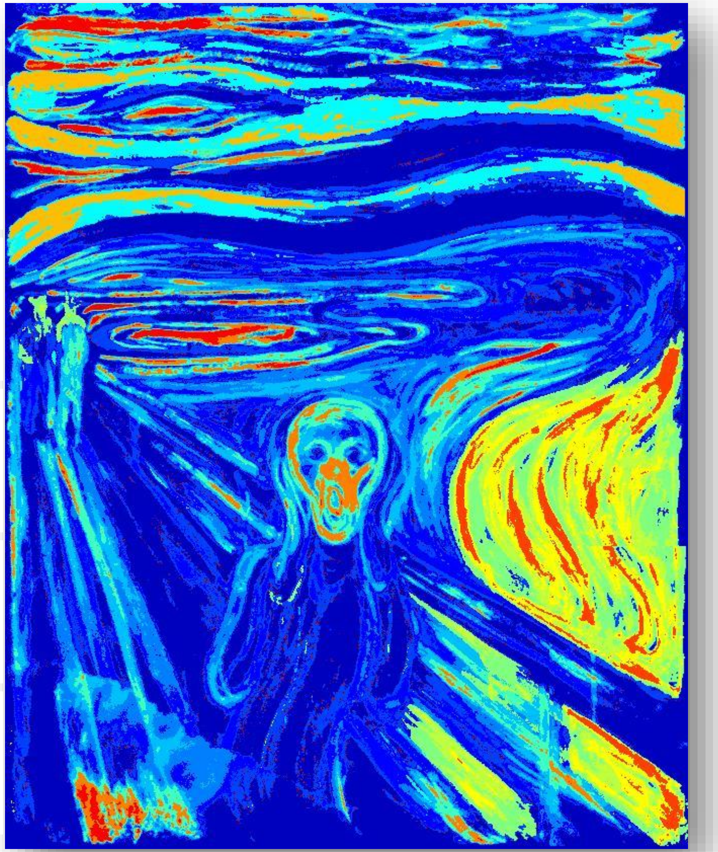
1. Partition of data in classes $C_i, i = 1, \dots, n$ using FCM;

# Two-stage FCM-based analysis

**FIRST STAGE:**

1. Partition of data in classes $C_i, i = 1, \dots, n$ using FCM;



# Two-stage FCM-based analysis

**FIRST STAGE:**

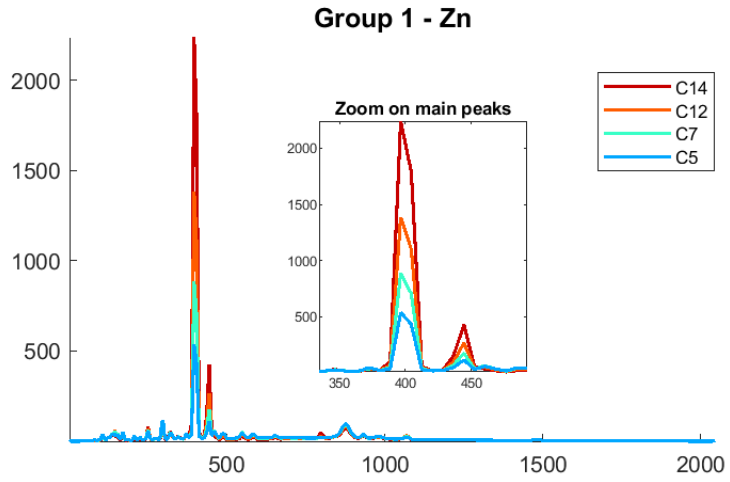
1. Partition of data in classes $C_i, i = 1, \dots, n$ using FCM;
2. Identification of classes of interest, and grouping according to the dominant peaks of the spectrum of the class center $c_i$.

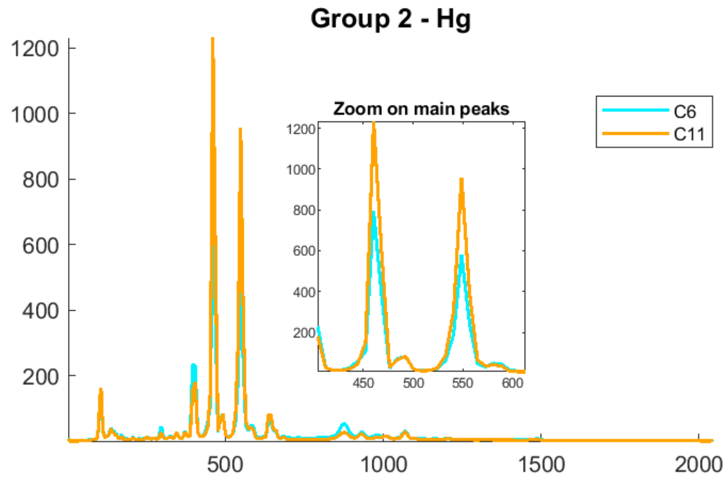
# Two-stage FCM-based analysis

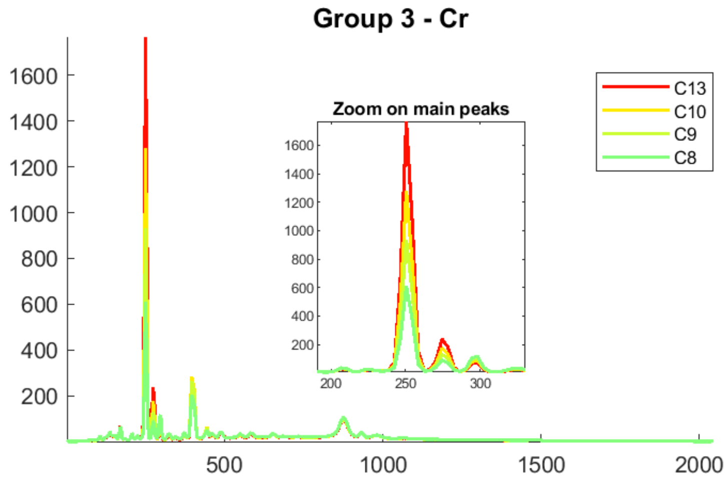
**FIRST STAGE:**

1. Partition of data in classes $C_i, i = 1, \dots, n$ using FCM;
2. Identification of classes of interest, and grouping according to the dominant peaks of the spectrum of the class center $c_i$.

**Classes sharing the same dominant element are assigned to the same group**



# Two-stage FCM-based analysis

**SECOND STAGE:** *iterative process*

*3.* For each group $G_j$, $j = 1, \dots, K$, and each class $C_i$ in $G_j$:

**While** predominant elements are detected and classes $C_i$ are separated from FCM:

*3.1.* Removal of predominant element peaks from pixels in $C_i$;

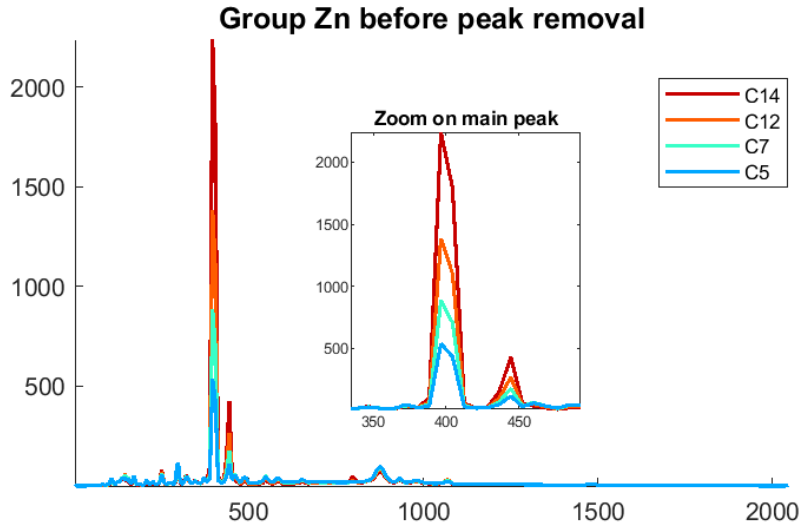
# Two-stage FCM-based analysis

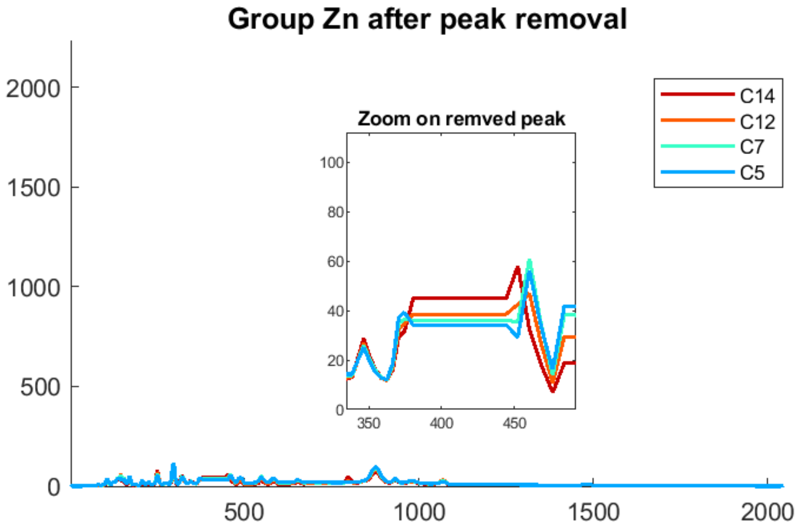
**SECOND STAGE:** *iterative process*

*3.* For each group $G_j$, $j = 1, \dots, K$, and each class $C_i$ in $G_j$:

**While** predominant elements are detected and classes $C_i$ are separated from FCM:

*3.1.* Removal of predominant element peaks from pixels in $C_i$;



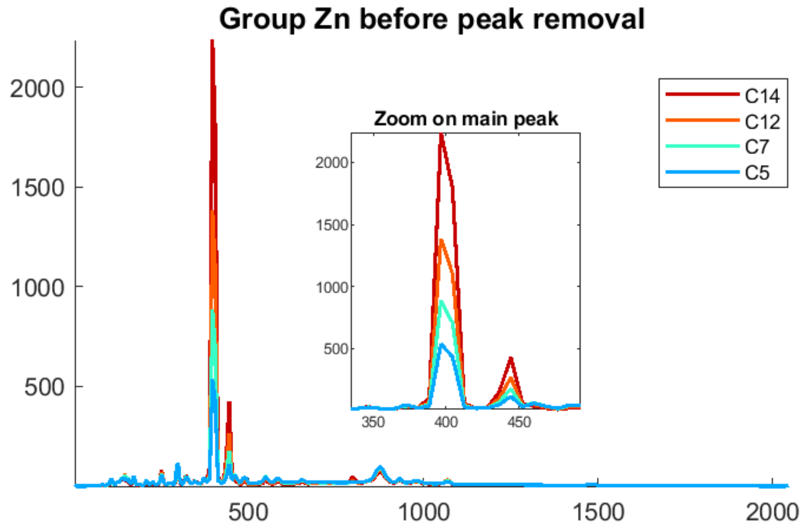
# Two-stage FCM-based analysis

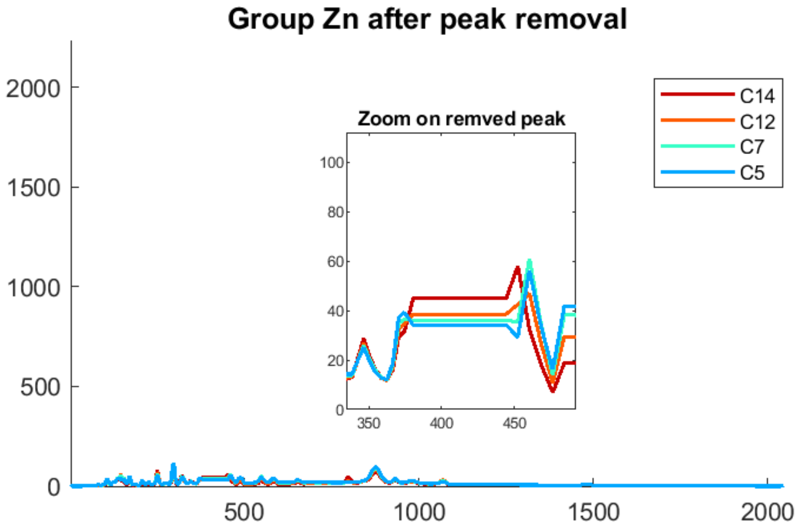
**SECOND STAGE:** *iterative process*

*3.* For each group $G_j$, $j = 1, \dots, K$, and each class $C_i$ in $G_j$:

**While** predominant elements are detected and classes $C_i$ are separated from FCM:

*3.1.* Removal of predominant element peaks from pixels in $C_i$;




*3.2.* Performing of FCM on $C_i$.

# Experimental settings

**DATA AVERAGING:** moving average filters

- 4 channels before 8 keV (~ first 380 channels)
- 8 channels after 8 keV

# Experimental settings

**DATA AVERAGING:** moving average filters

- 4 channels before 8 keV (~ first 380 channels)
- 8 channels after 8 keV

**CLASS MERGING**

SECOND STAGE: small number of pixels in each class $C_i$, with low variability

→ merging operation based on the distance between the centers of the classes

# Experimental settings

**DATA AVERAGING:** moving average filters

- 4 channels before 8 keV (~ first 380 channels)
- 8 channels after 8 keV

**CLASS MERGING**

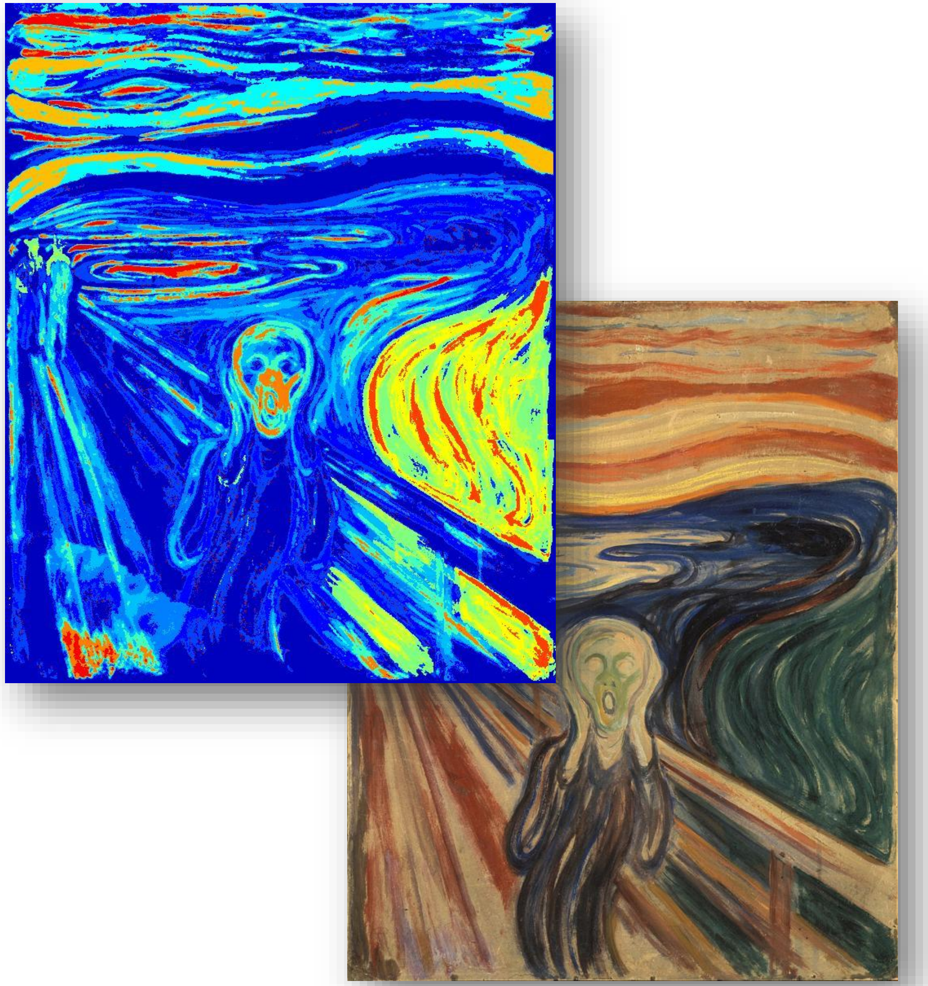
SECOND STAGE: small number of pixels in each class $C_i$, with low variability

→ merging operation based on the distance between the centers of the classes

**ALGORITHM SETTINGS**

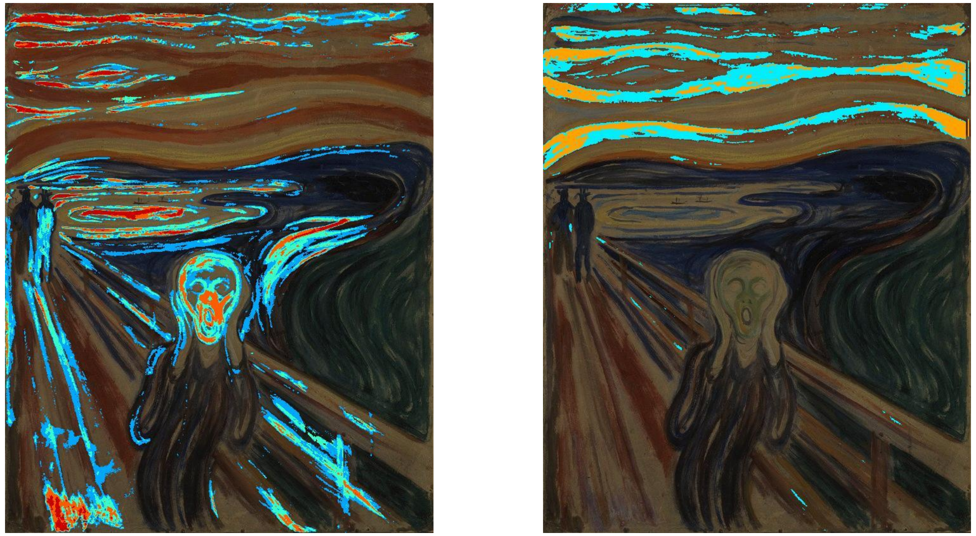
- Standard setting for the FCM parameters
- 15 classes used as FCM input at first stage

# Results - first stage



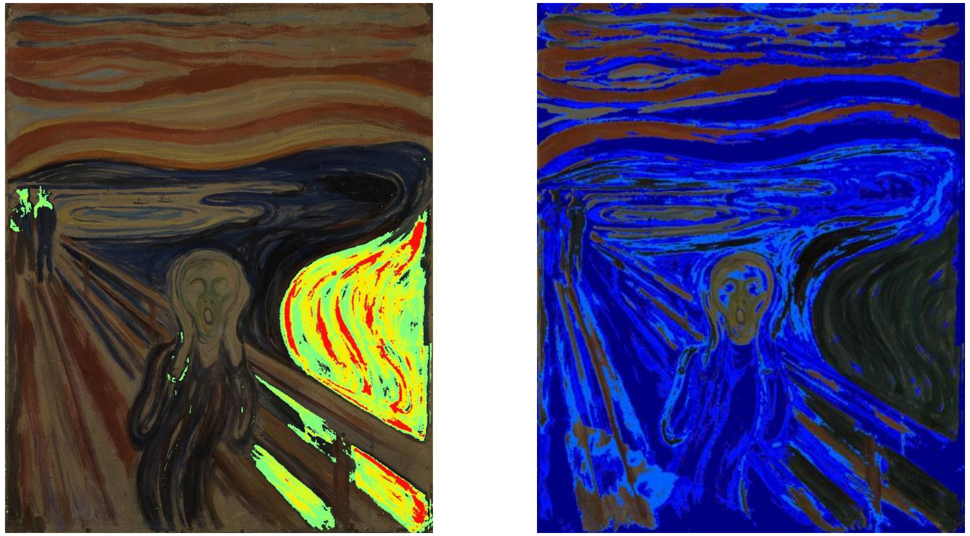
## Group 1 - Zinc

## Group 2 - Mercury



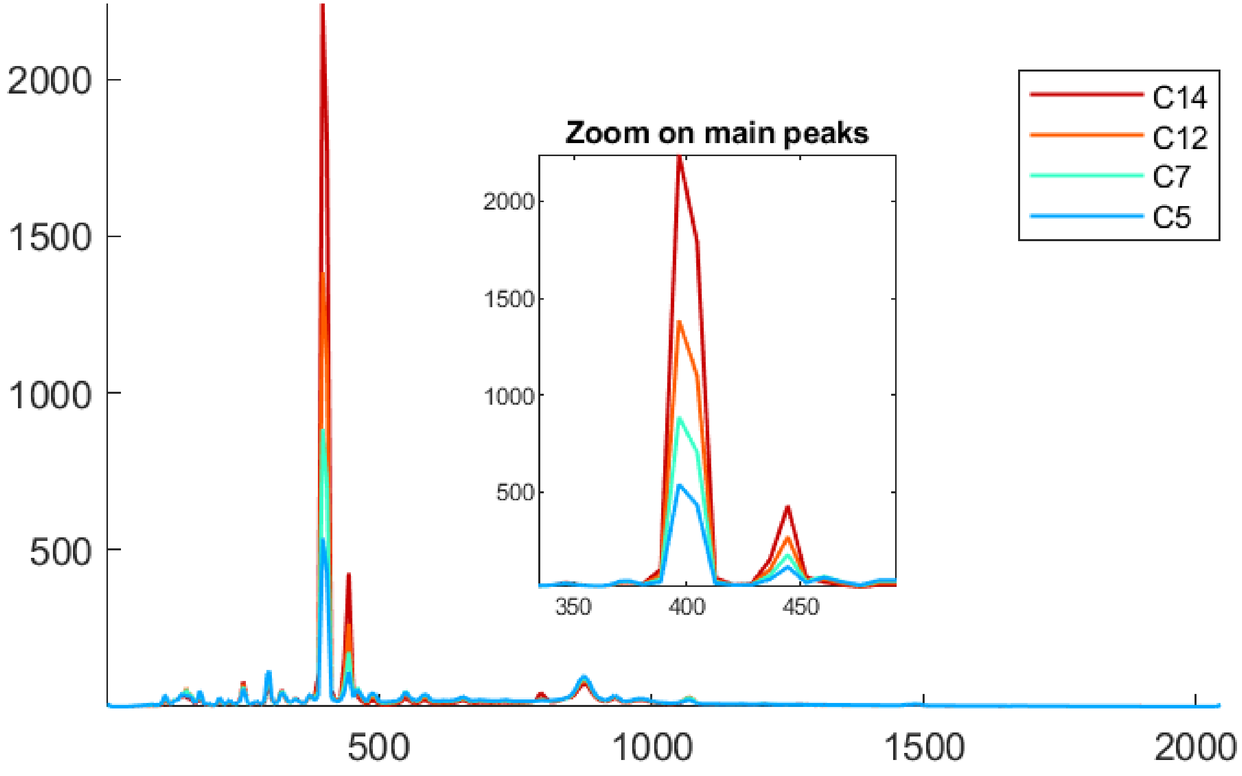
## Group 3 - Chrome

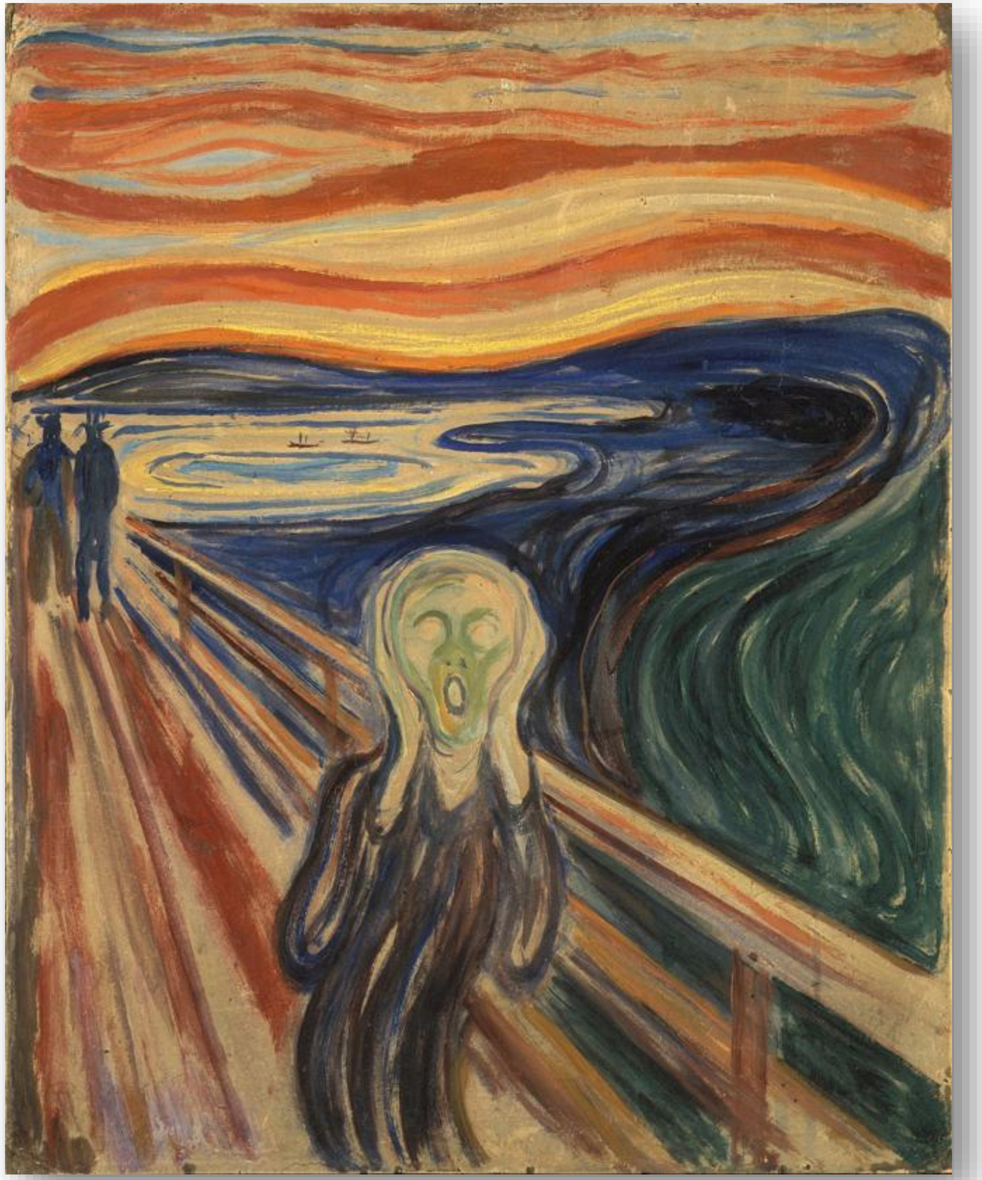
## Group 4 - Low signals



# Results - first stage

## Group 1 - Zinc

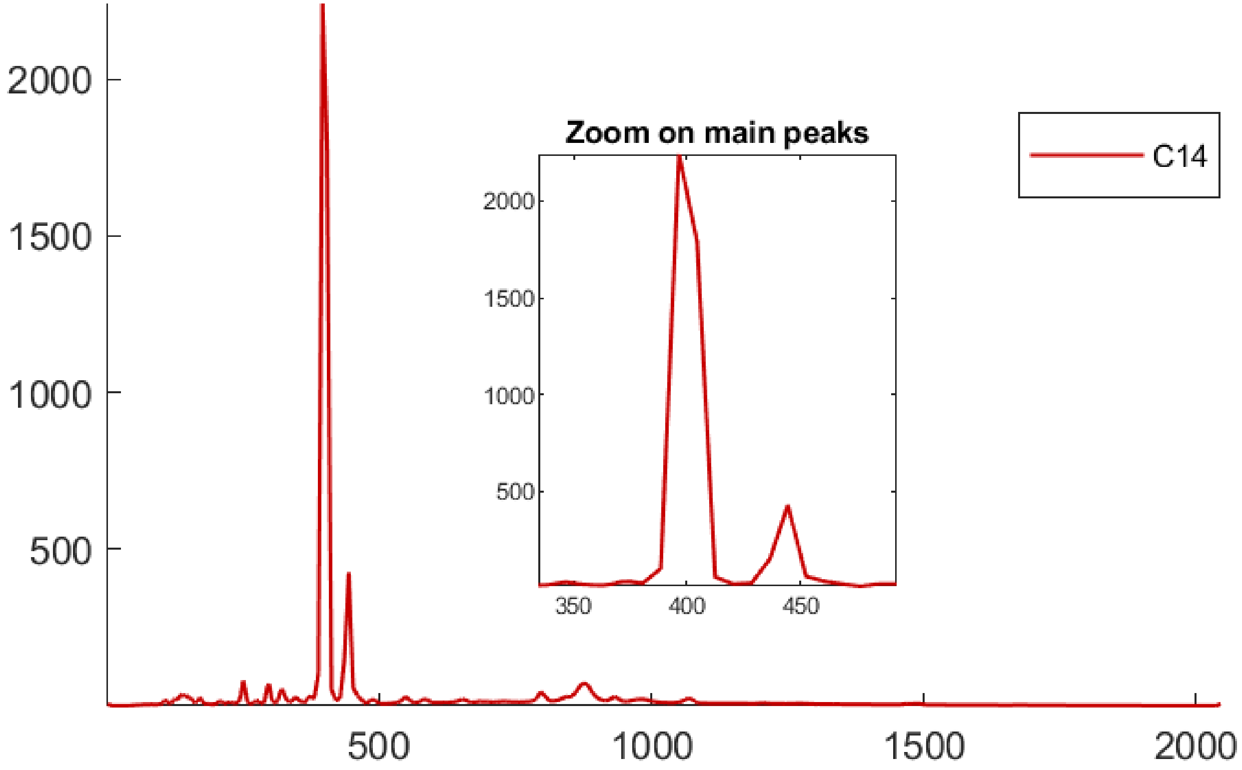


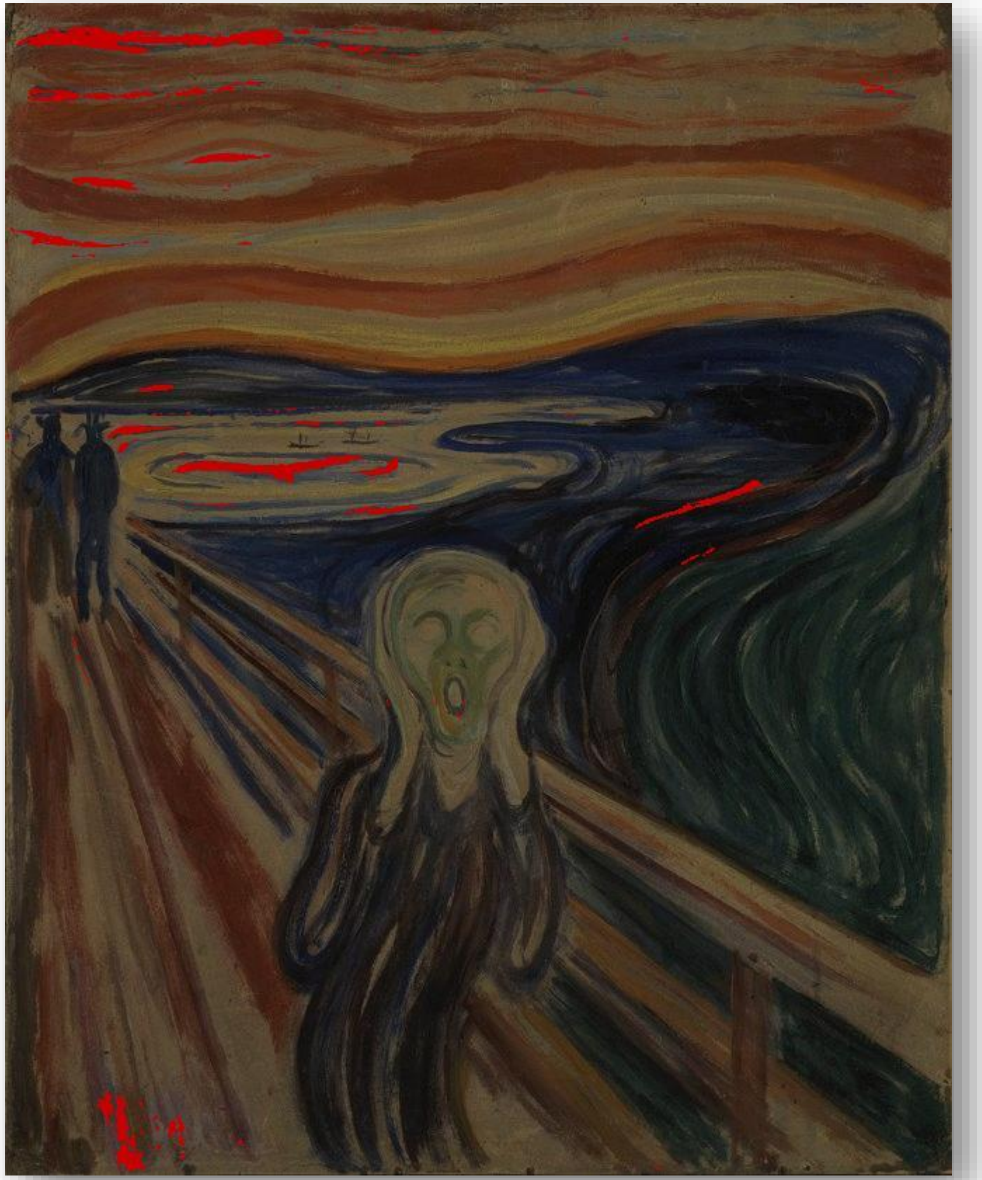



# Results - first stage

Zn +++

## Group 1 - Zinc



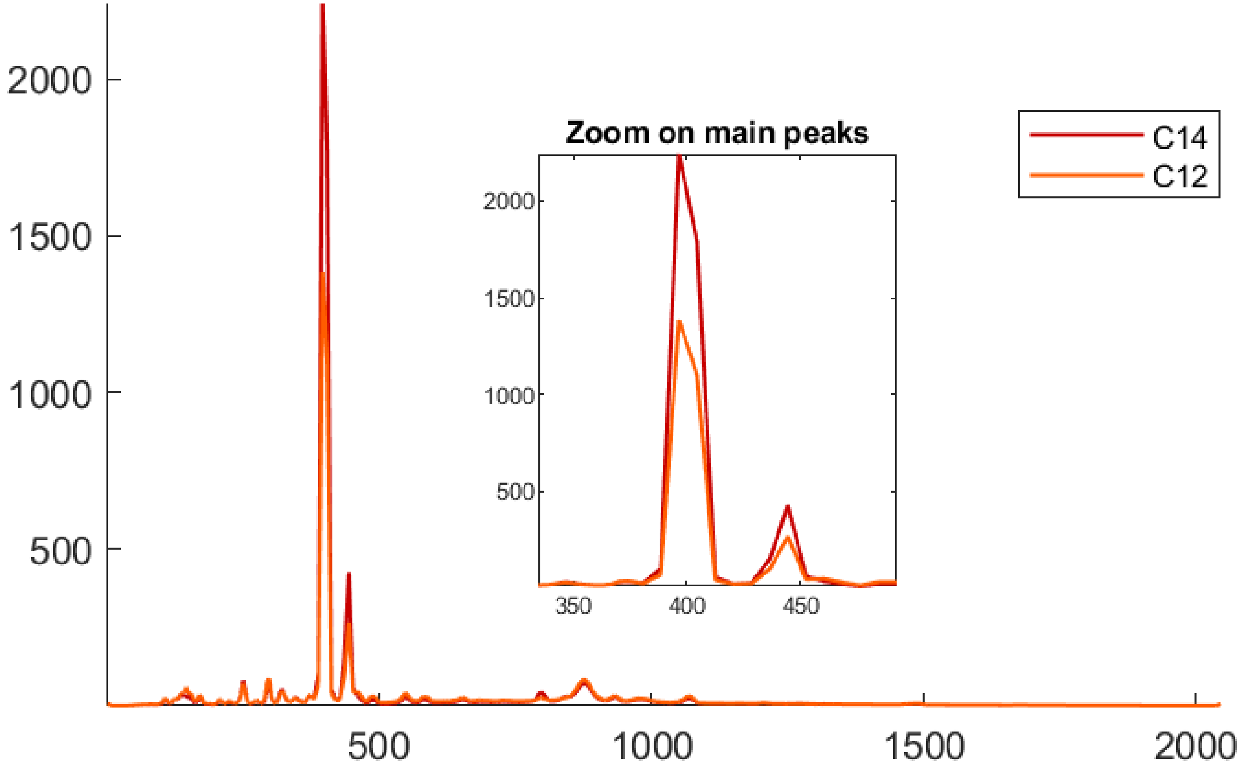



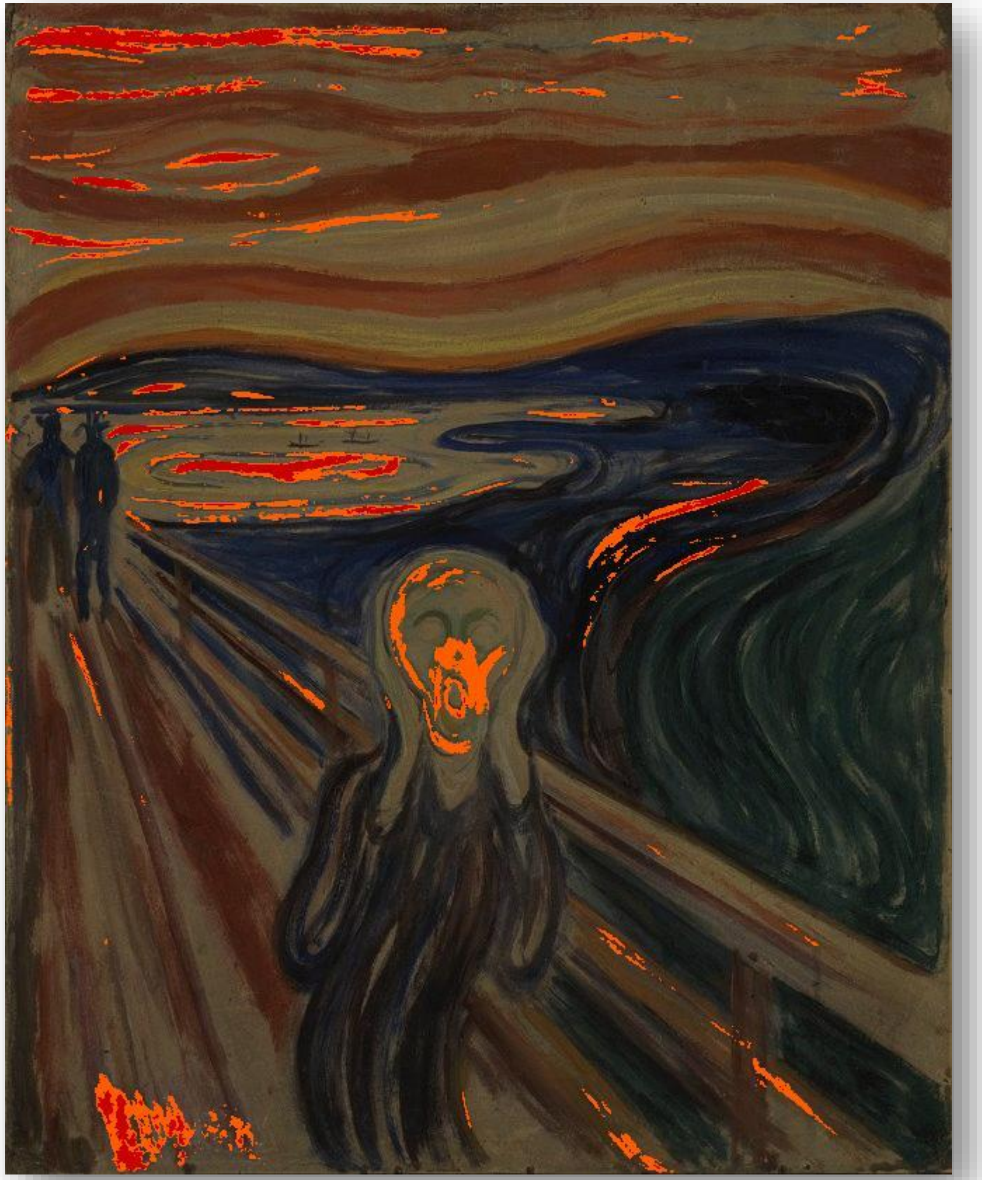
# Results - first stage

## Group 1 - Zinc

Zn +++

Zn ++

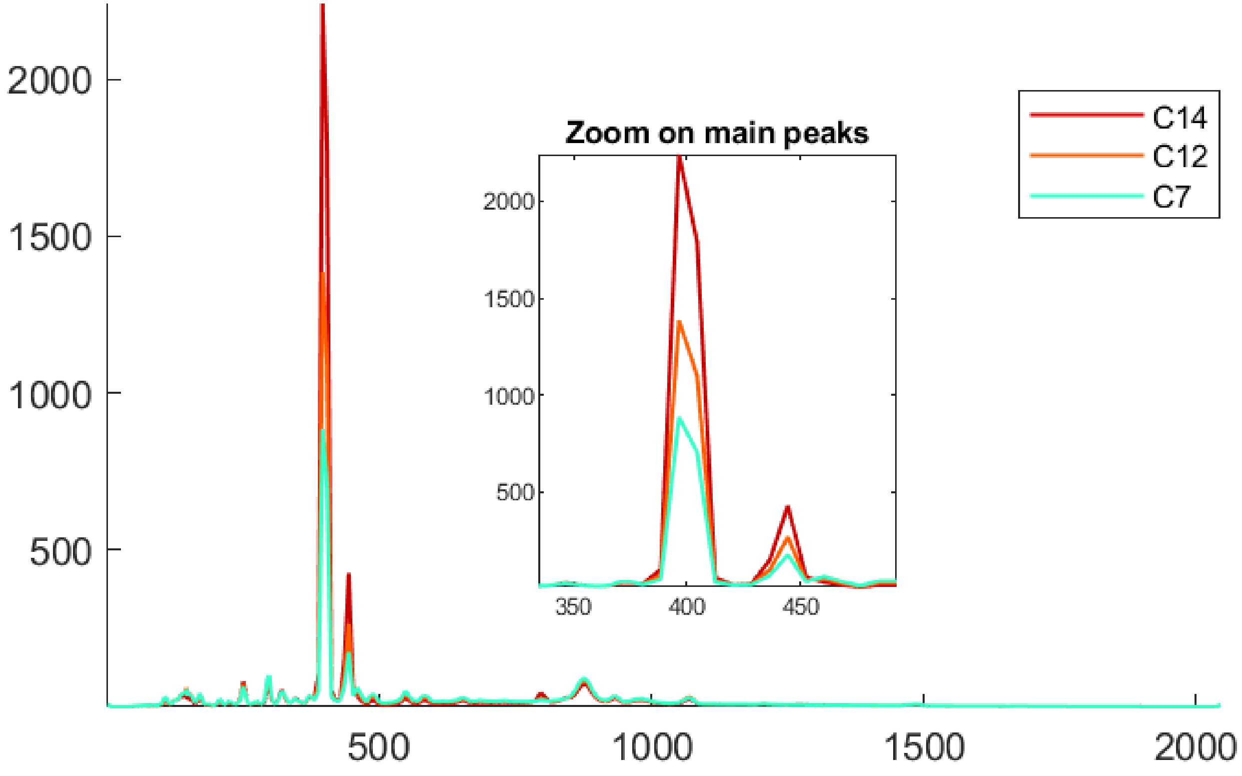


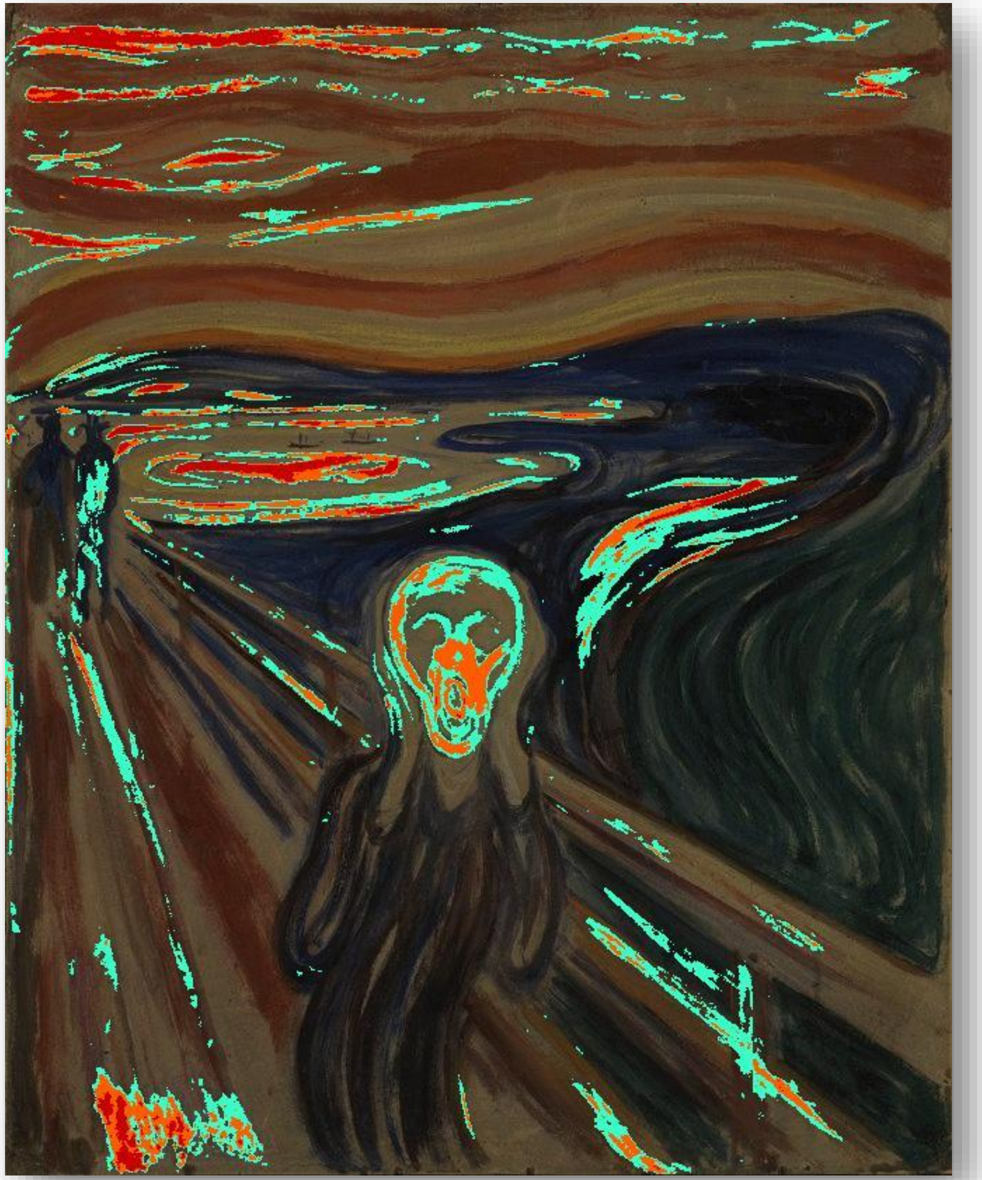



# Results - first stage

## Group 1 - Zinc

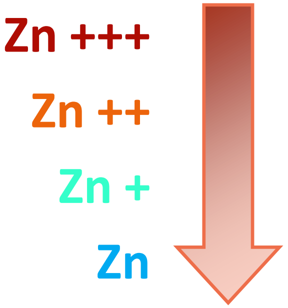
Zn +++
Zn ++
Zn +

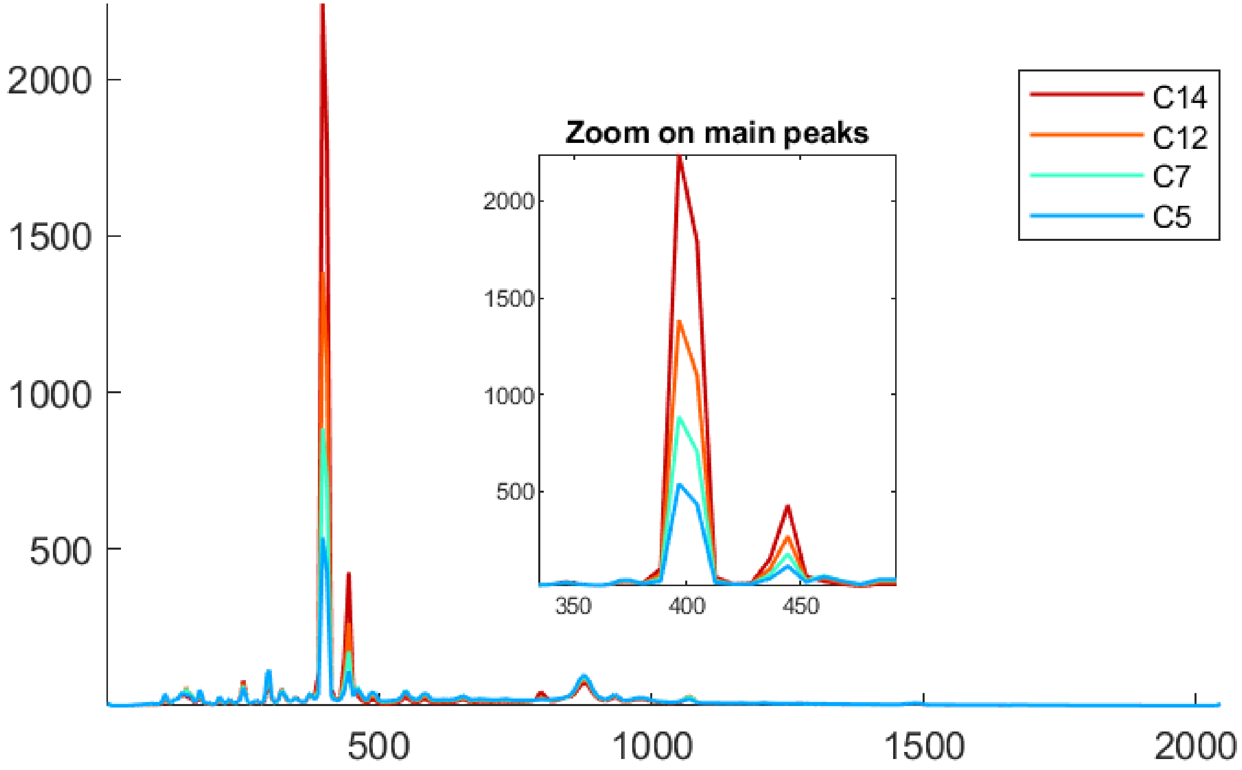


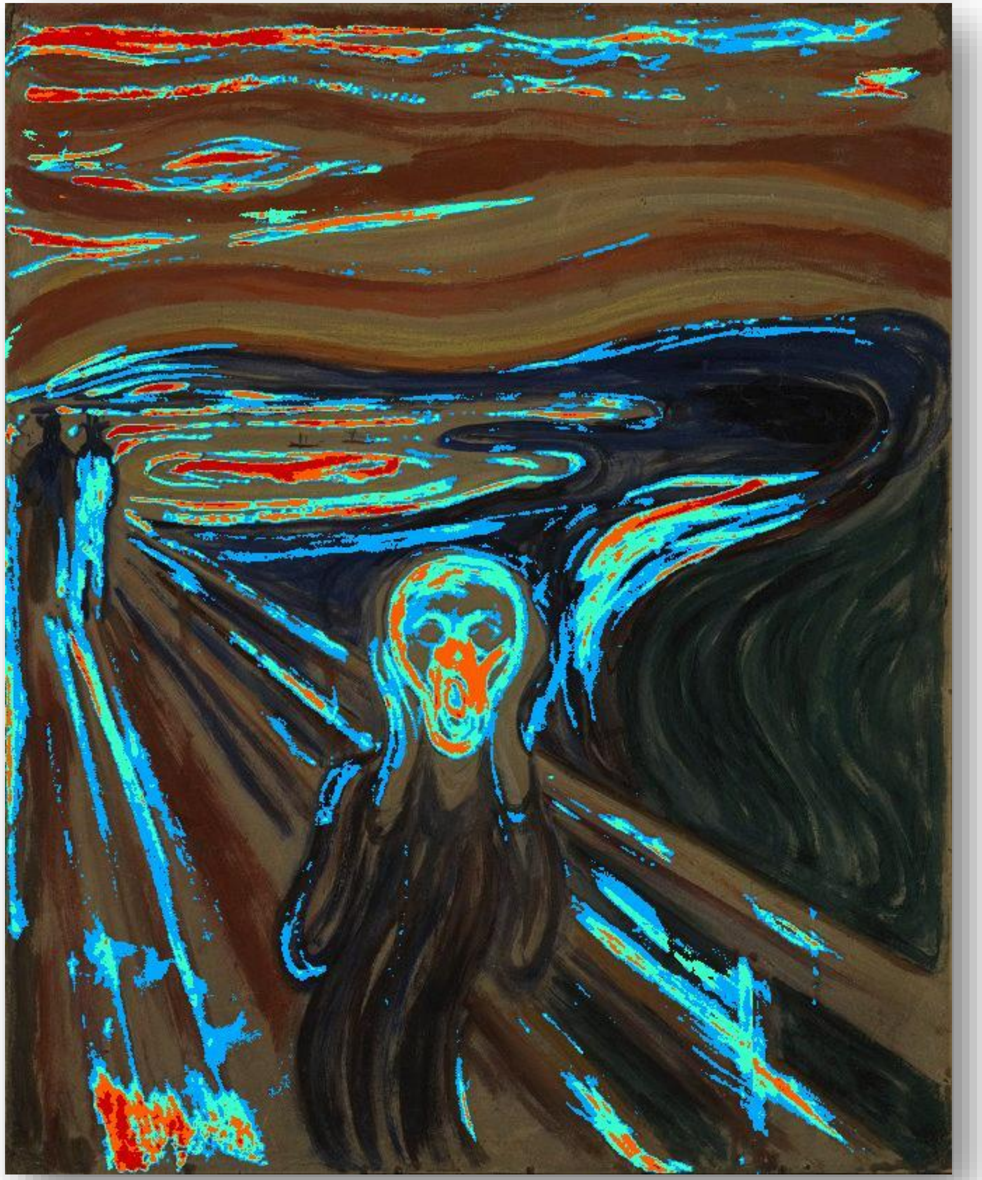



# Results - first stage

## Group 1 - Zinc

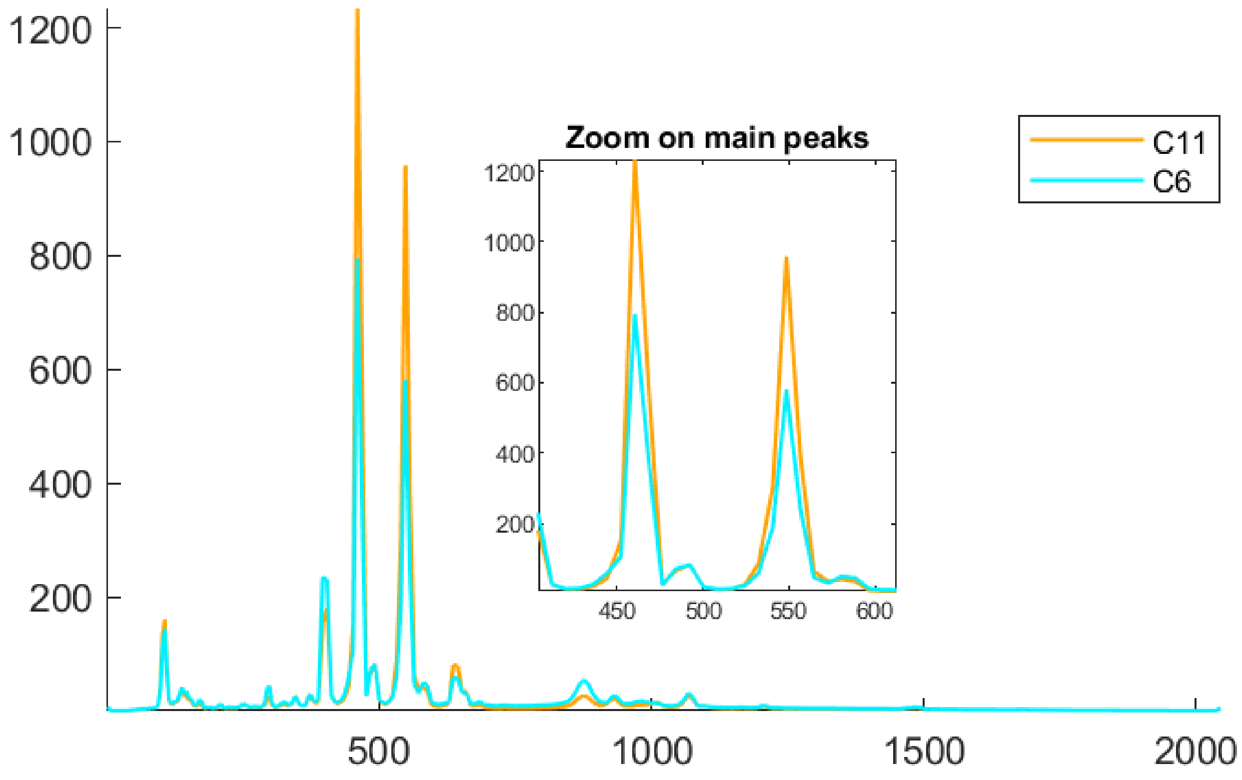


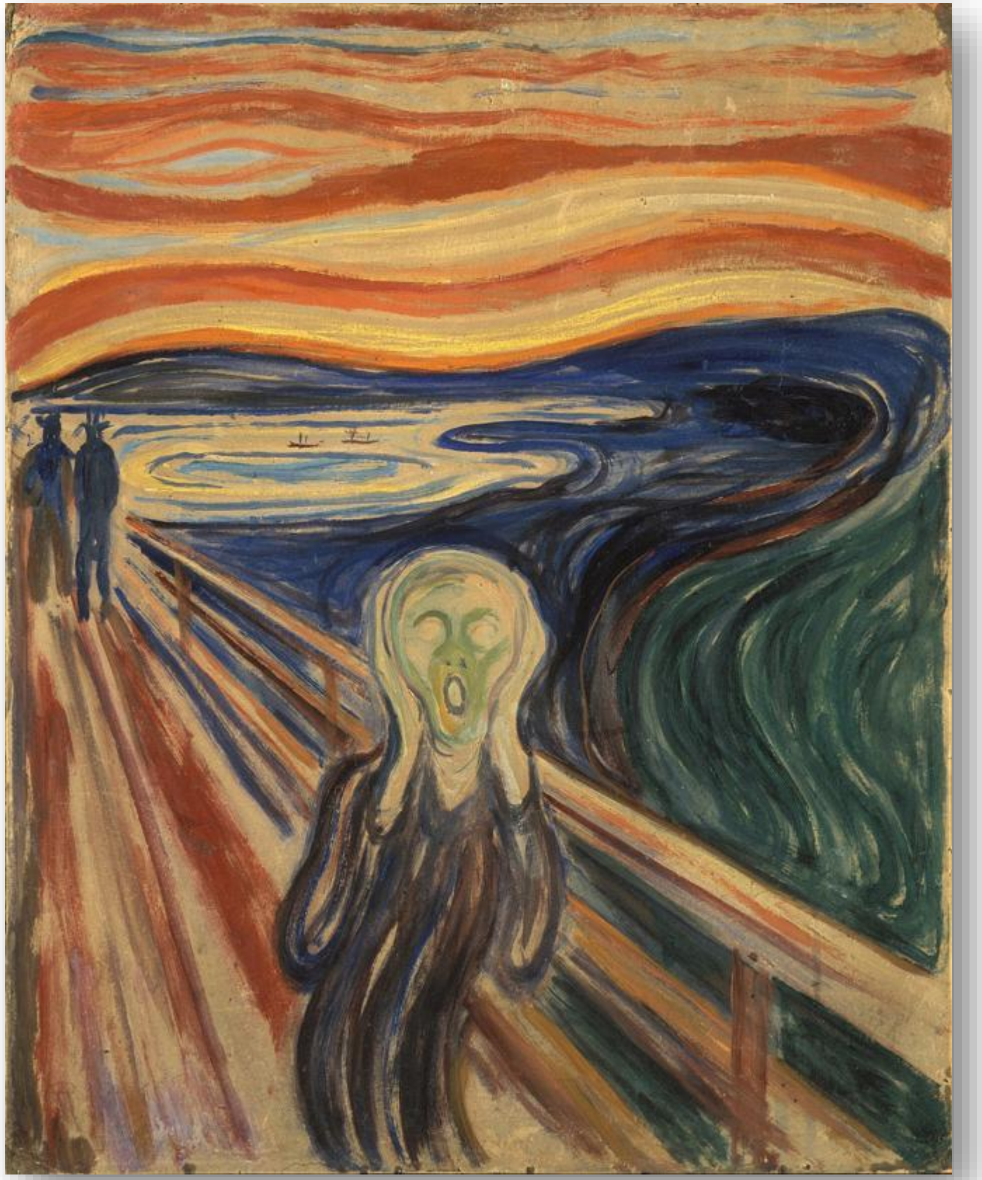



# Results - first stage

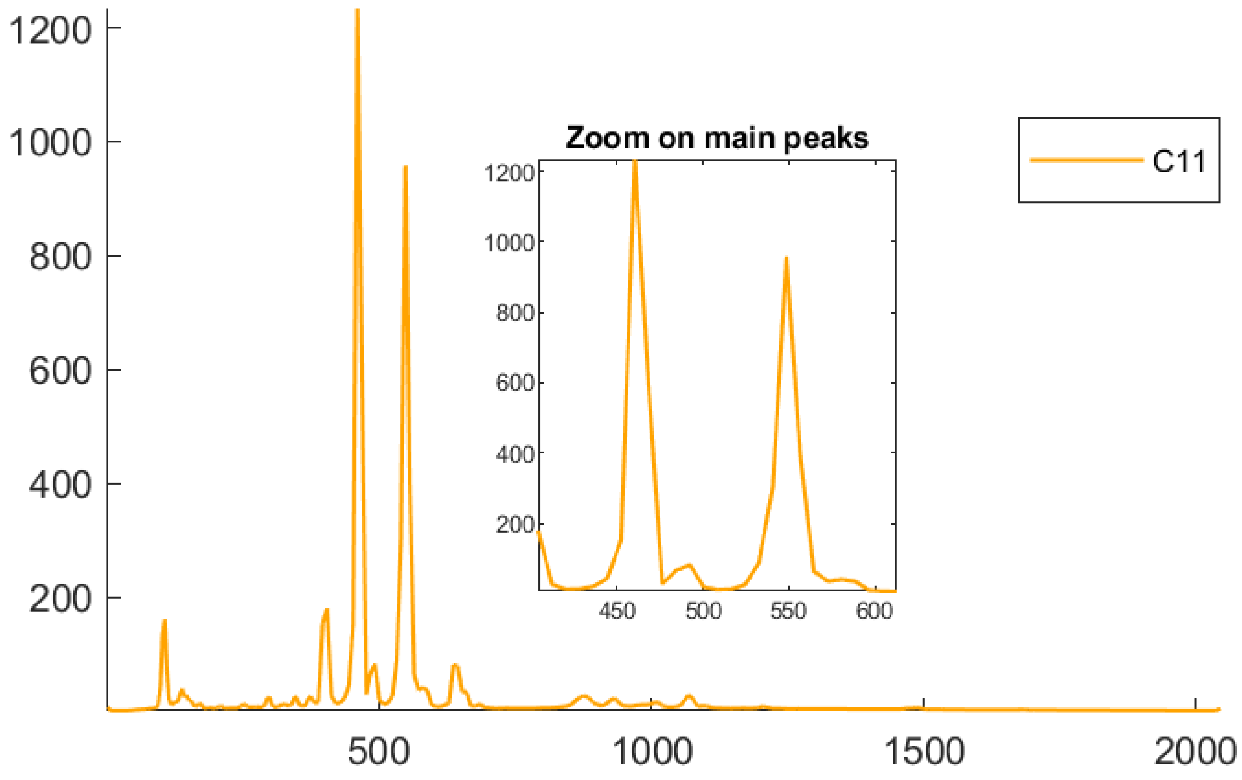
## Group 2 - Mercury

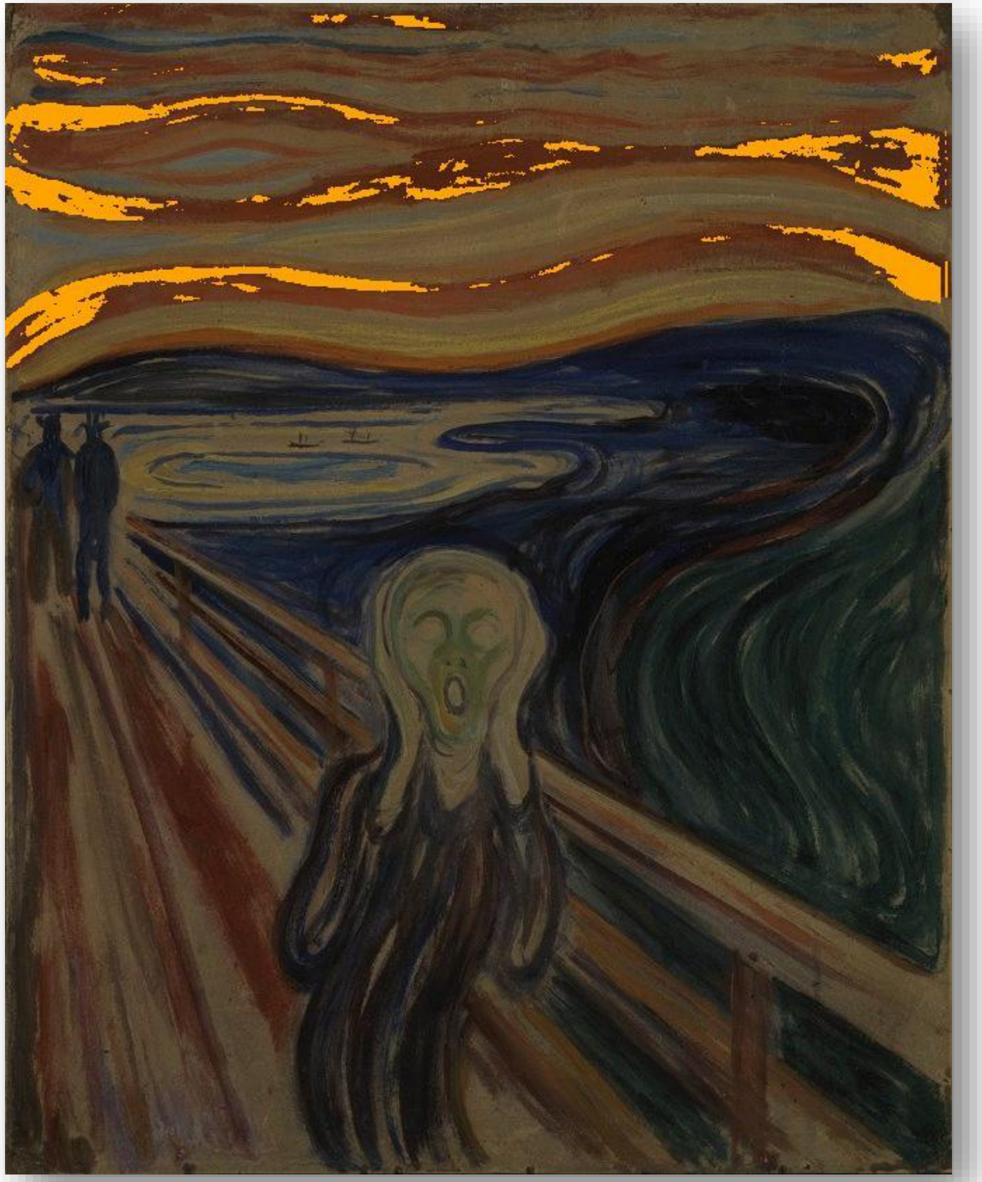


# Results - first stage

## Group 2 - Mercury



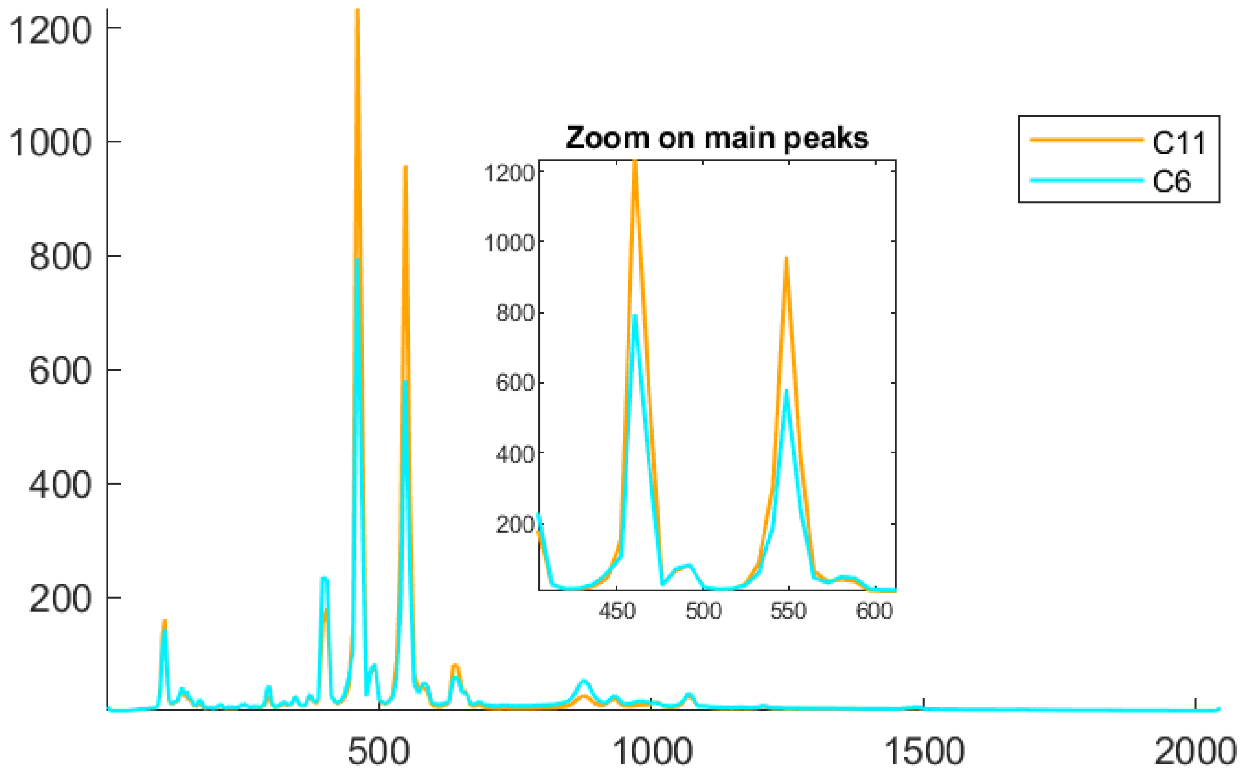



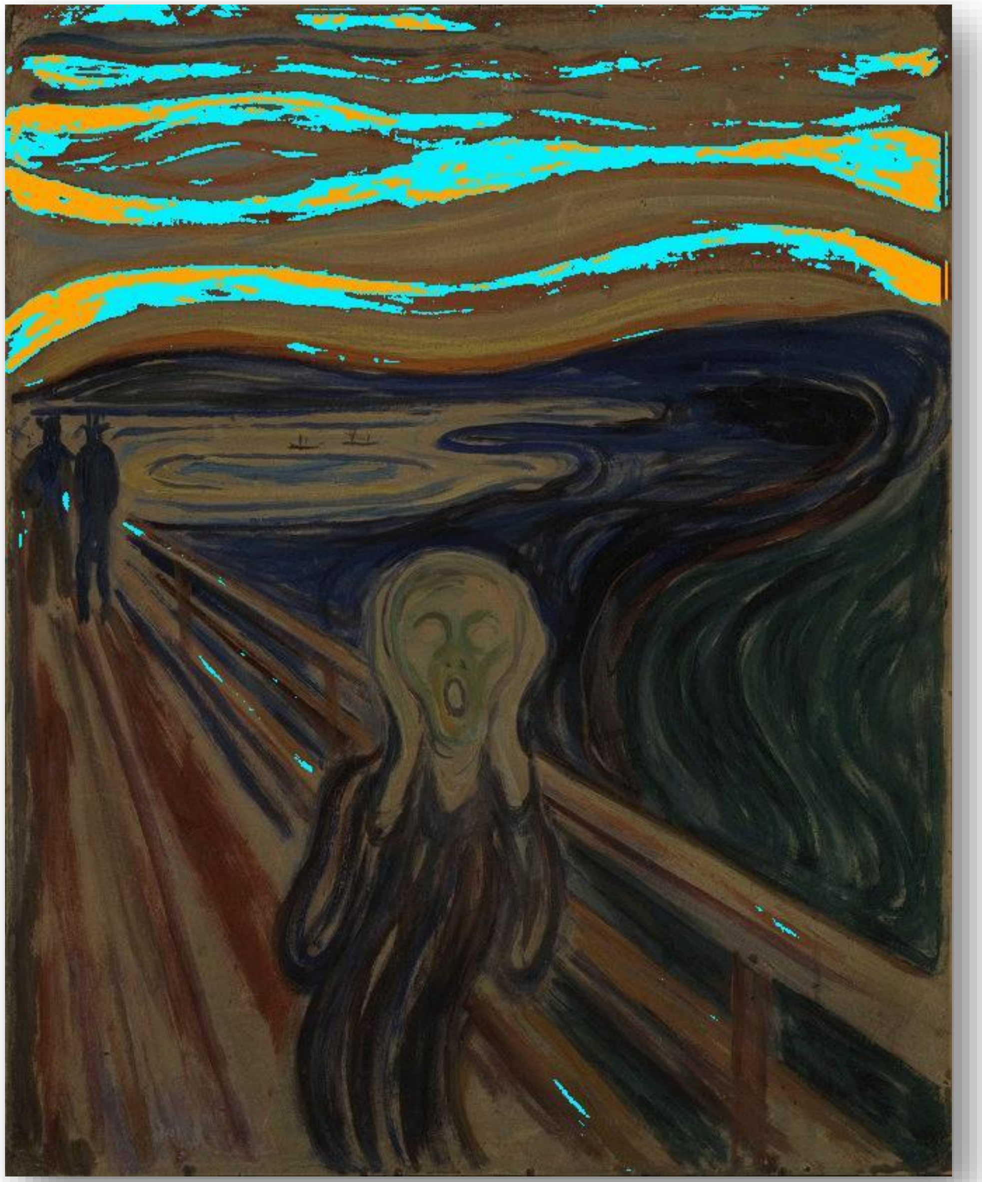
# Results - first stage

Hg +

Hg

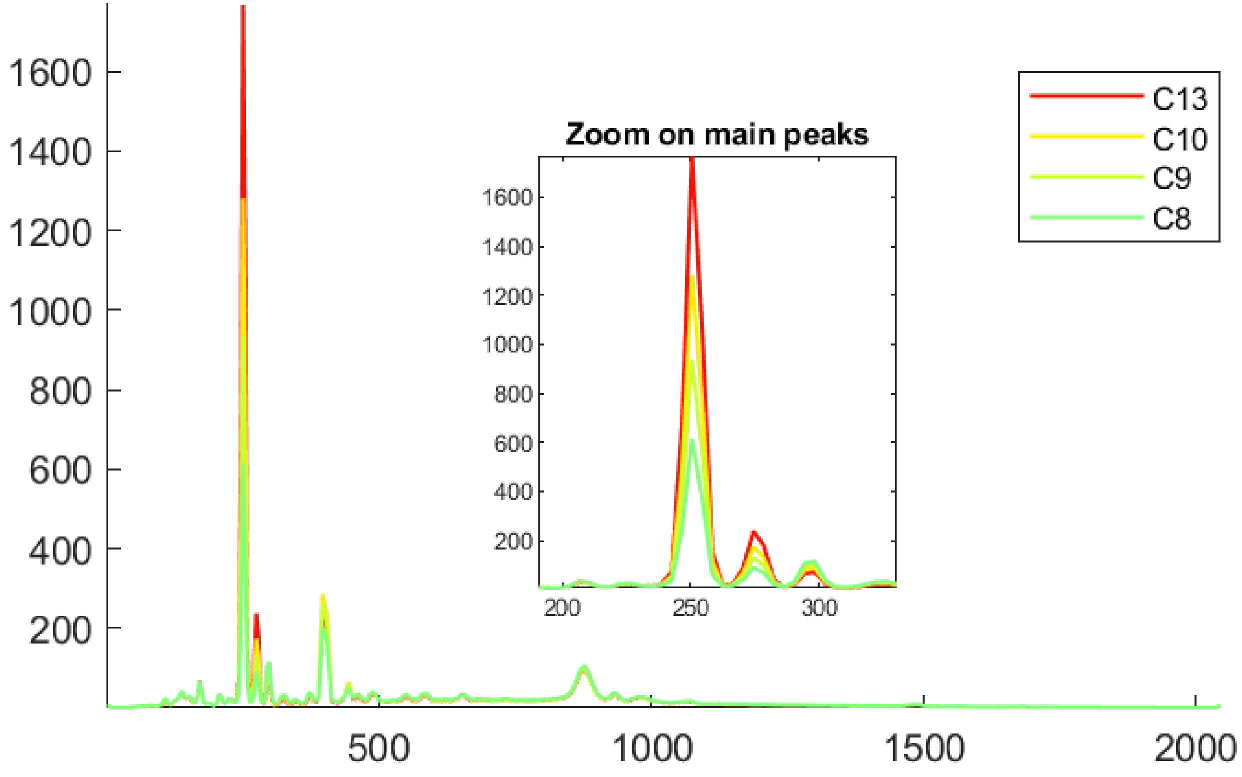
## Group 2 - Mercury

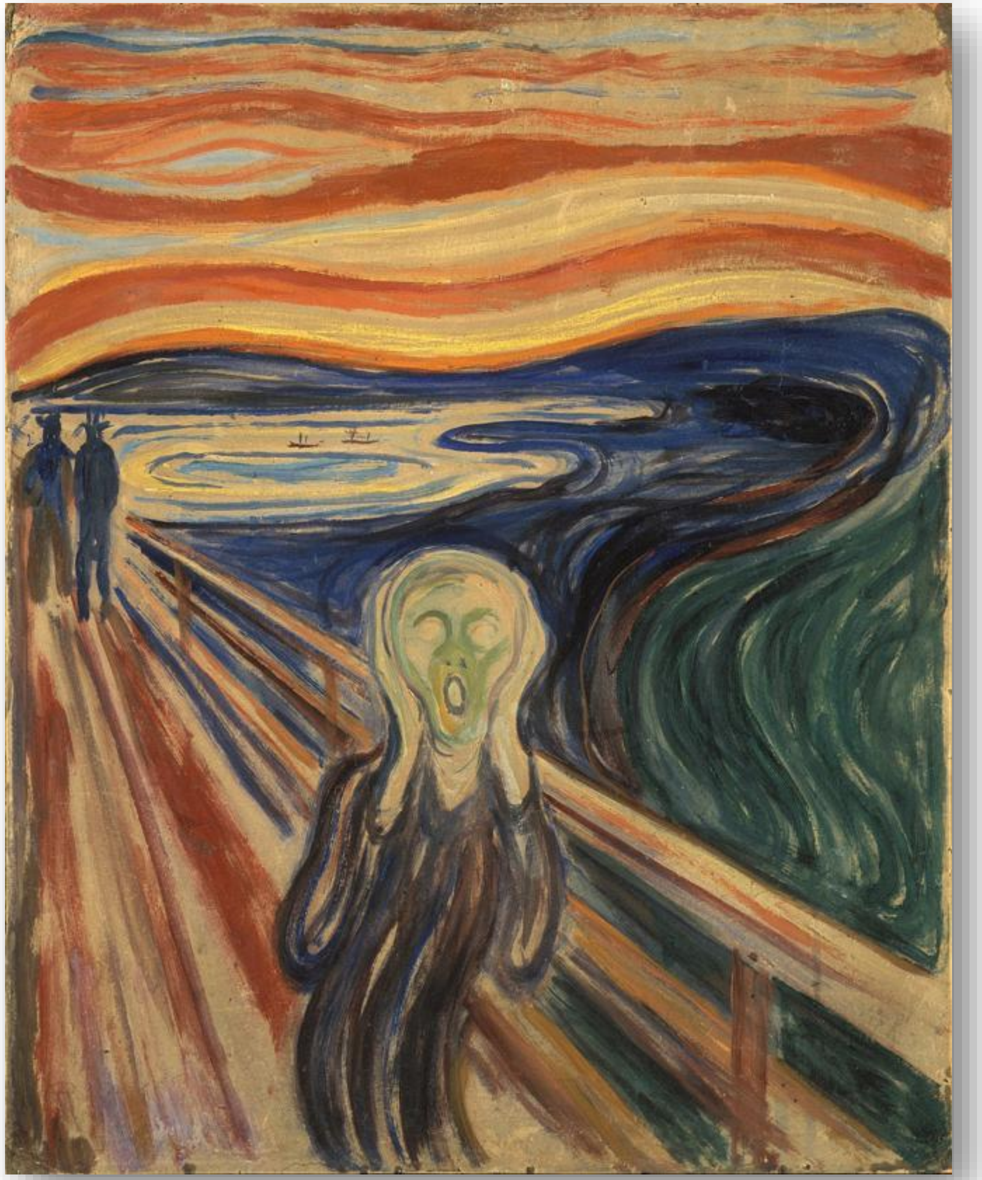


# Results - first stage

## Group 3 - Chrome

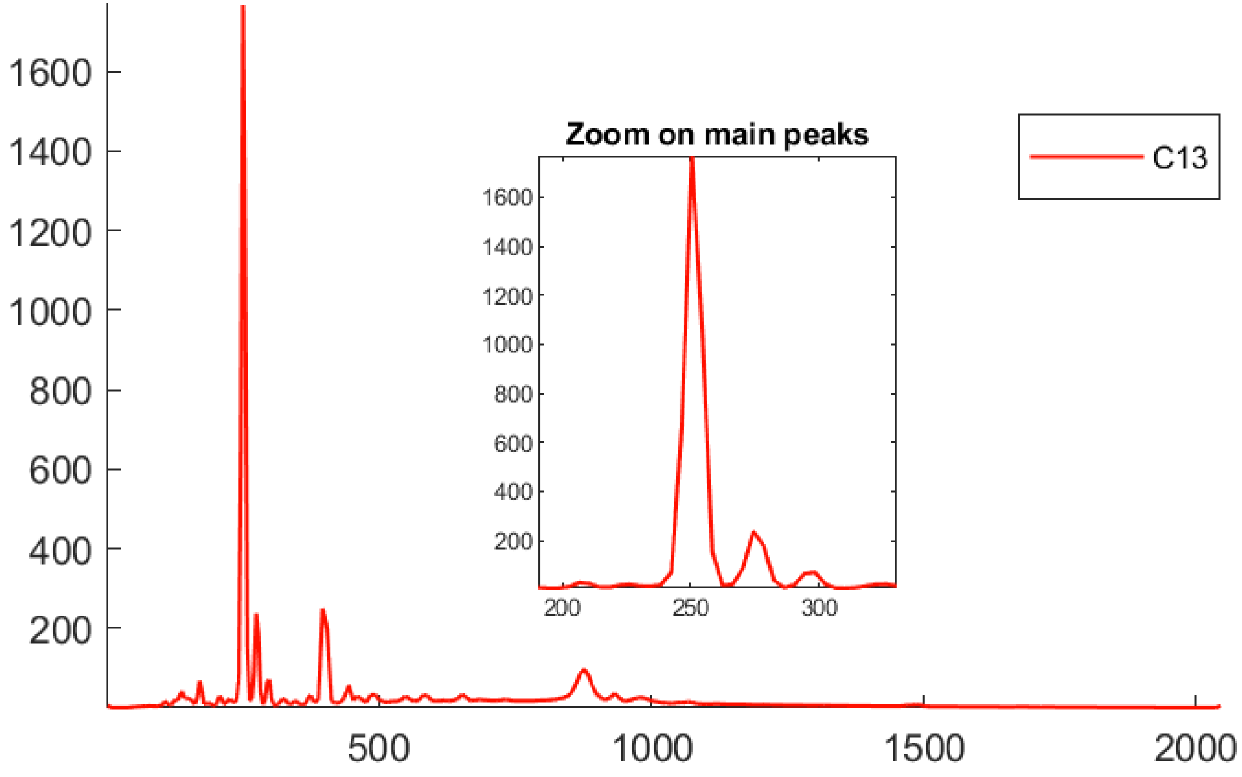


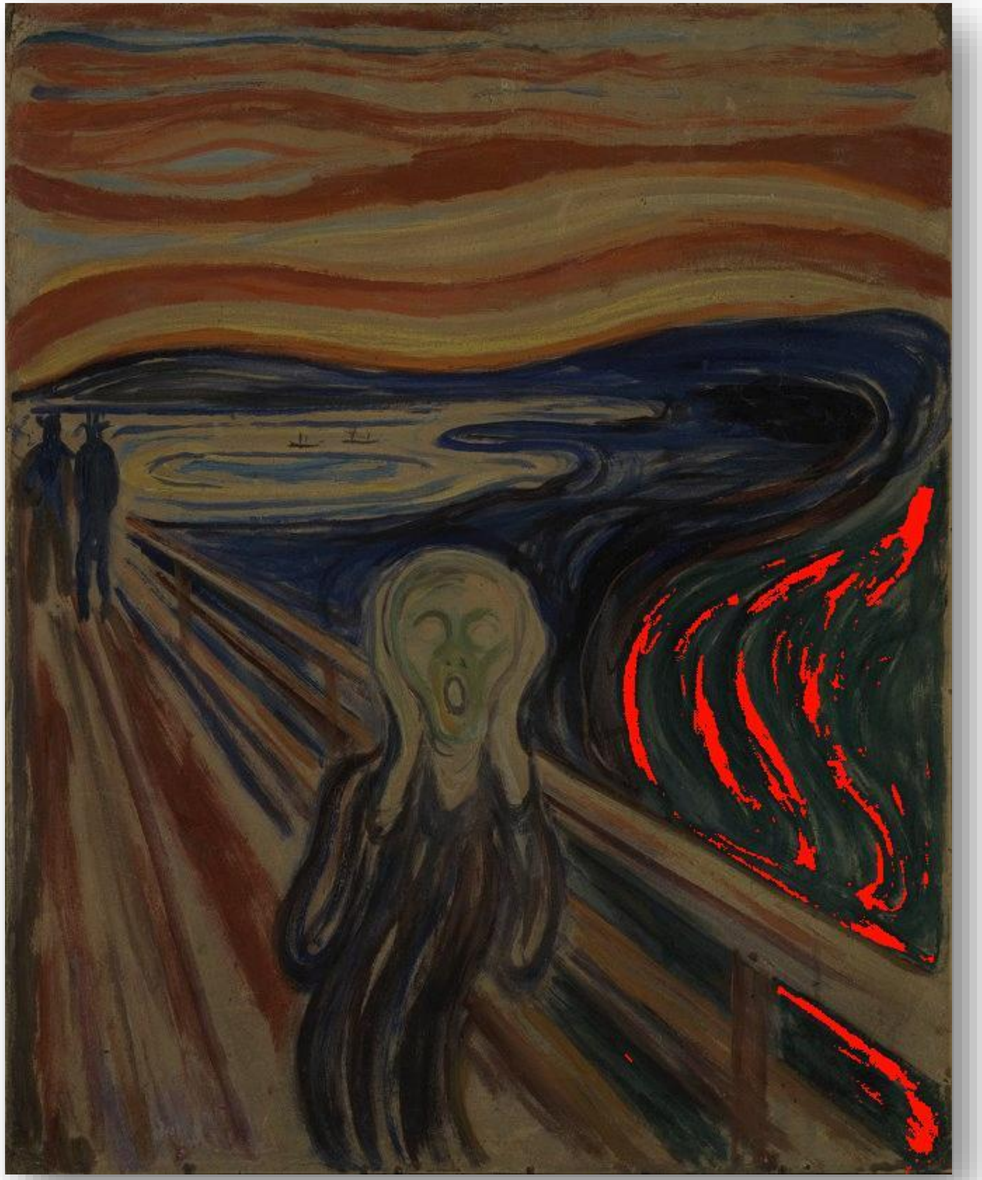



# Results - first stage

## Group 3 - Chrome

Cr +++



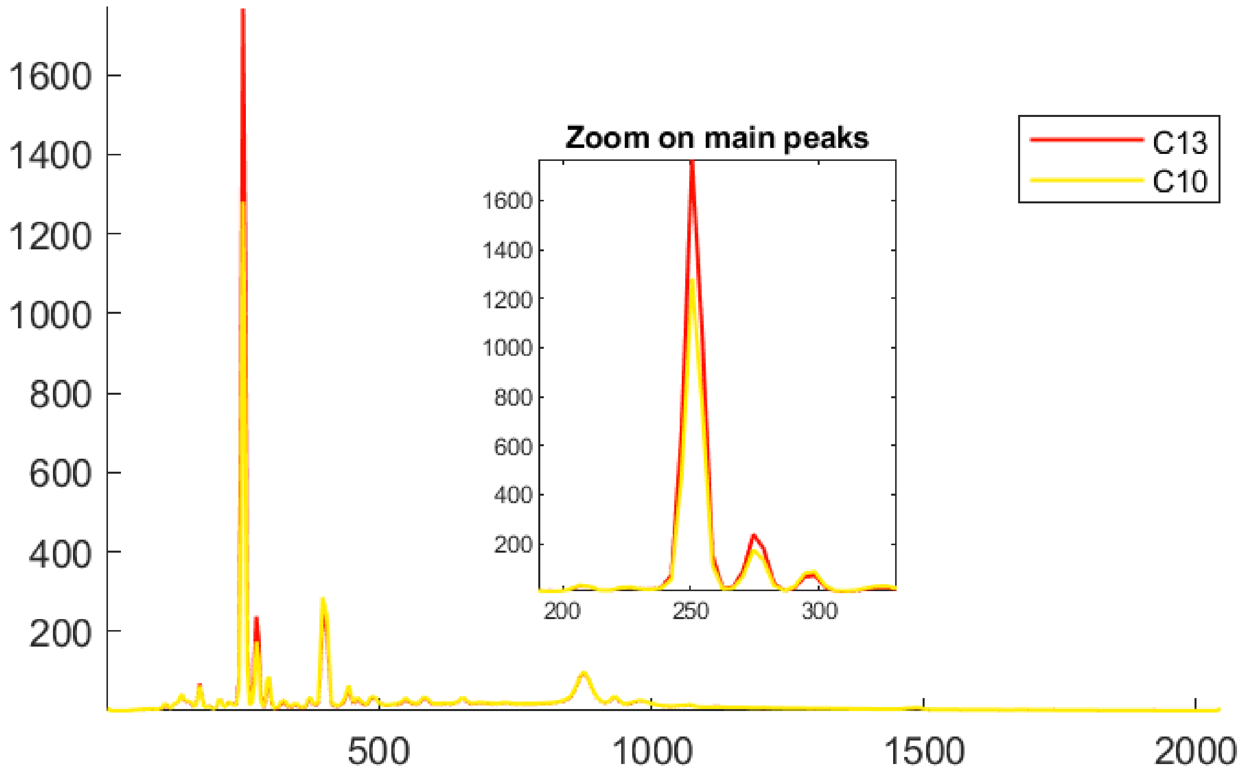



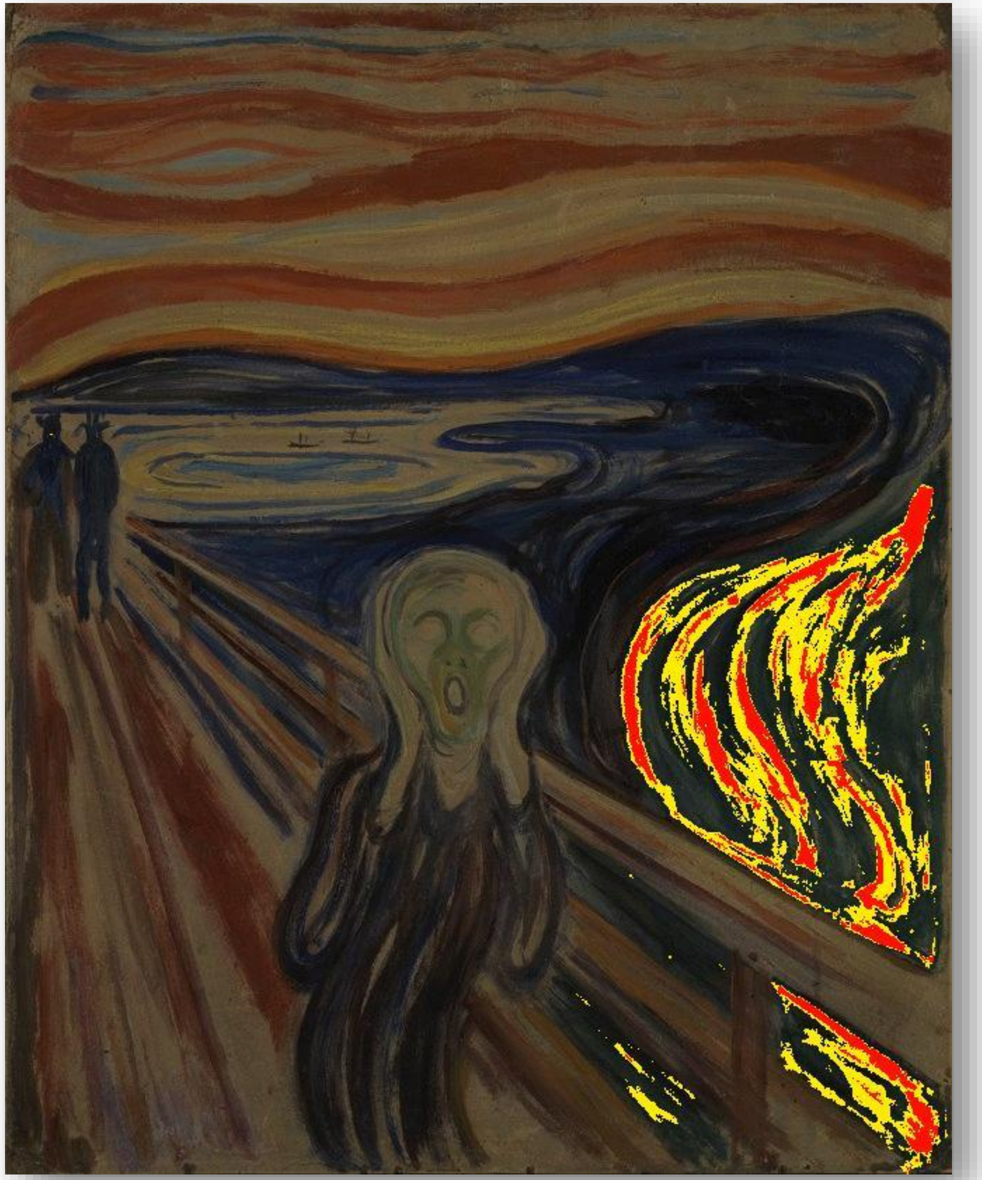
# Results - first stage

## Group 3 - Chrome

Cr +++

Cr ++

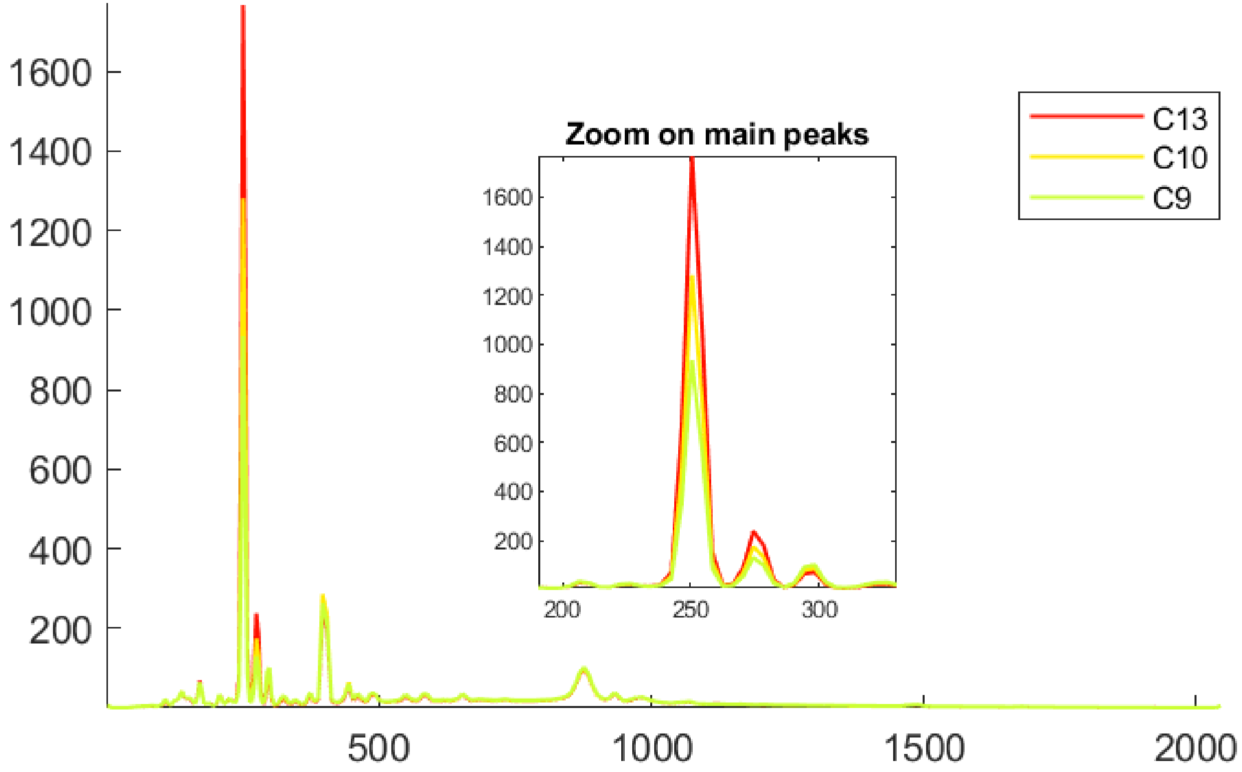


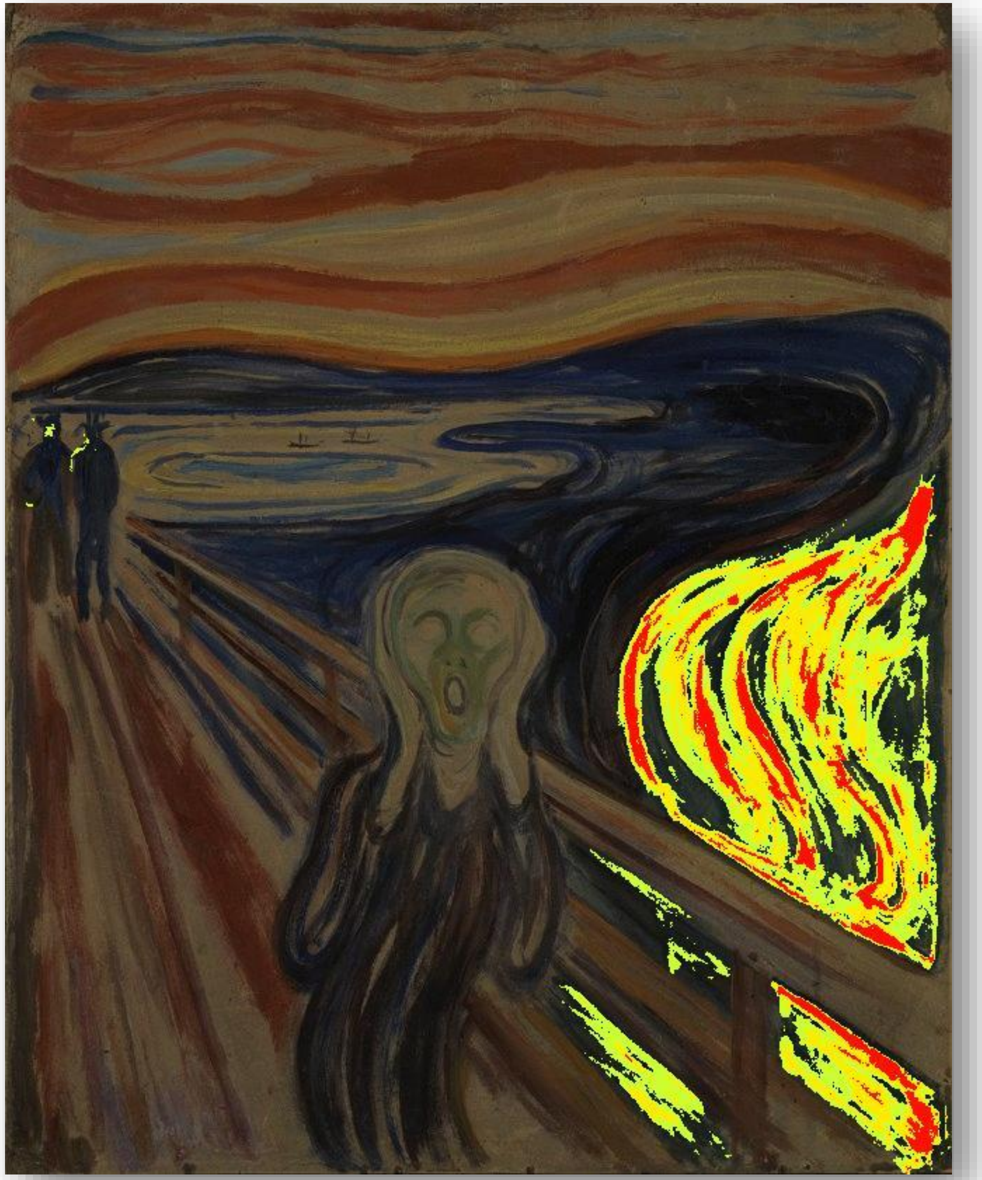



# Results - first stage

## Group 3 - Chrome

Cr +++
Cr ++
Cr +

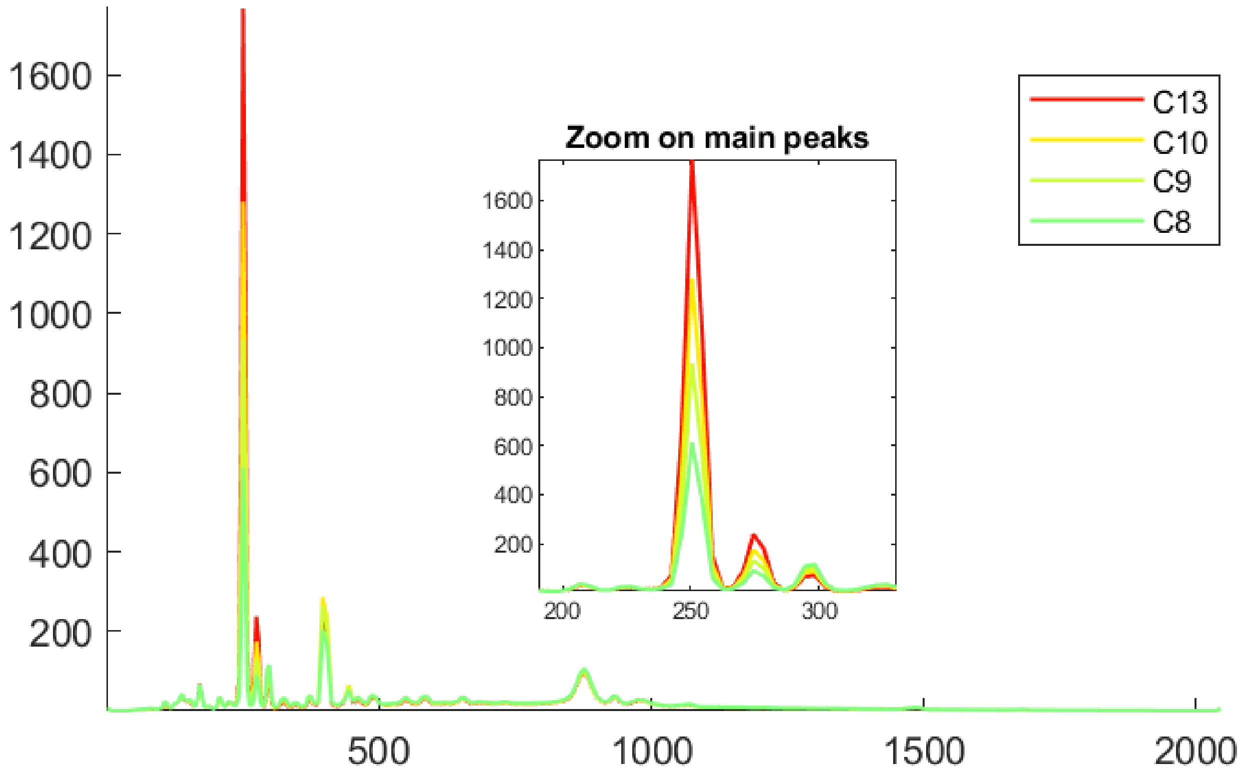


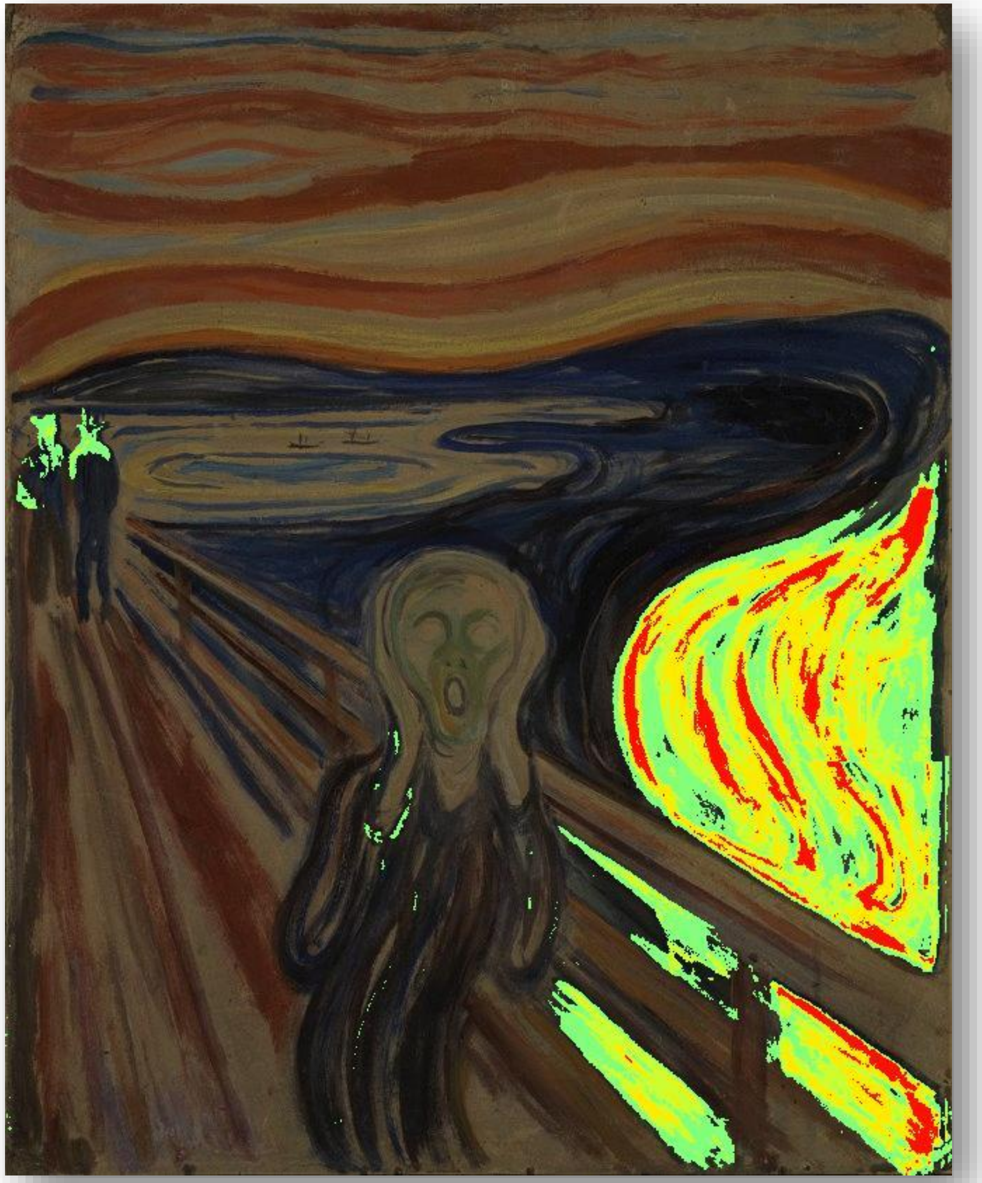



# Results - first stage

## Group 3 - Chrome

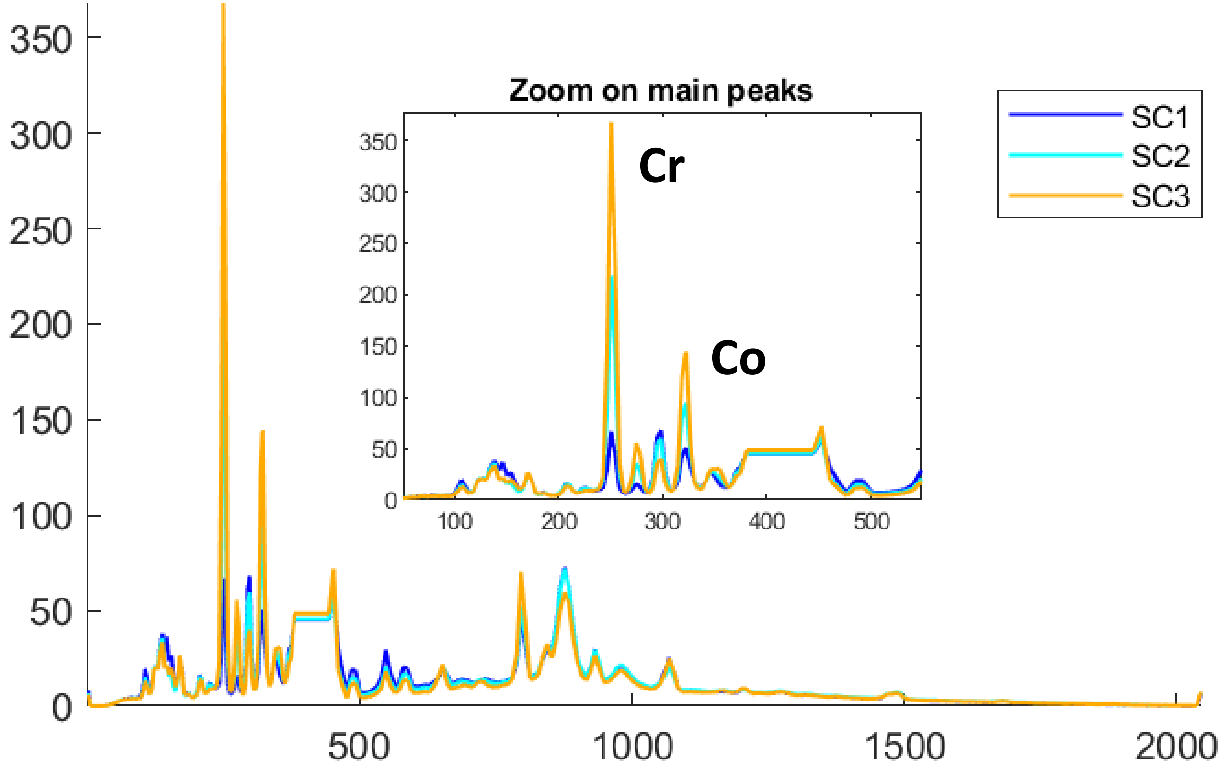
Cr +++
Cr ++
Cr +
Cr

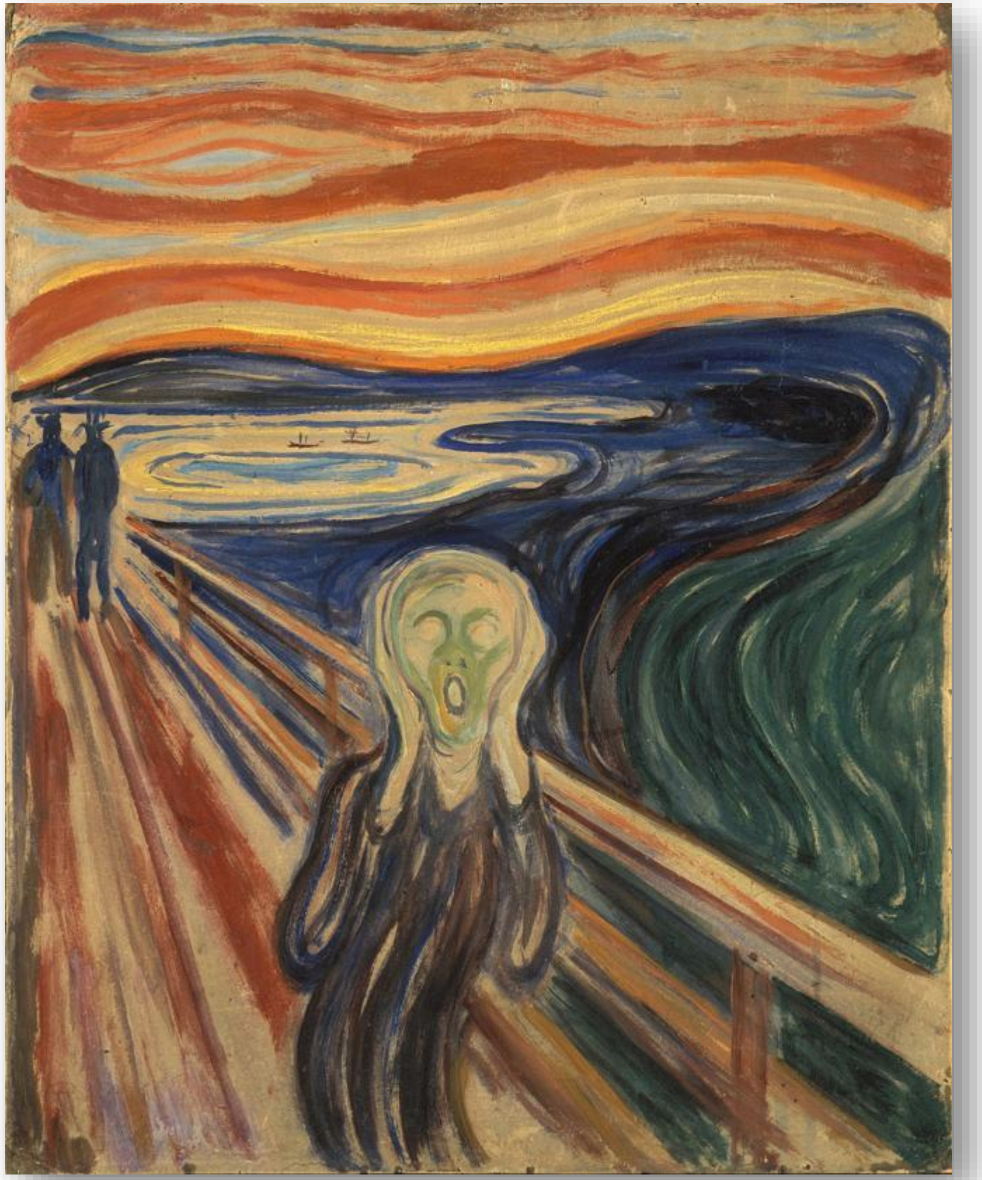


# Results - second stage

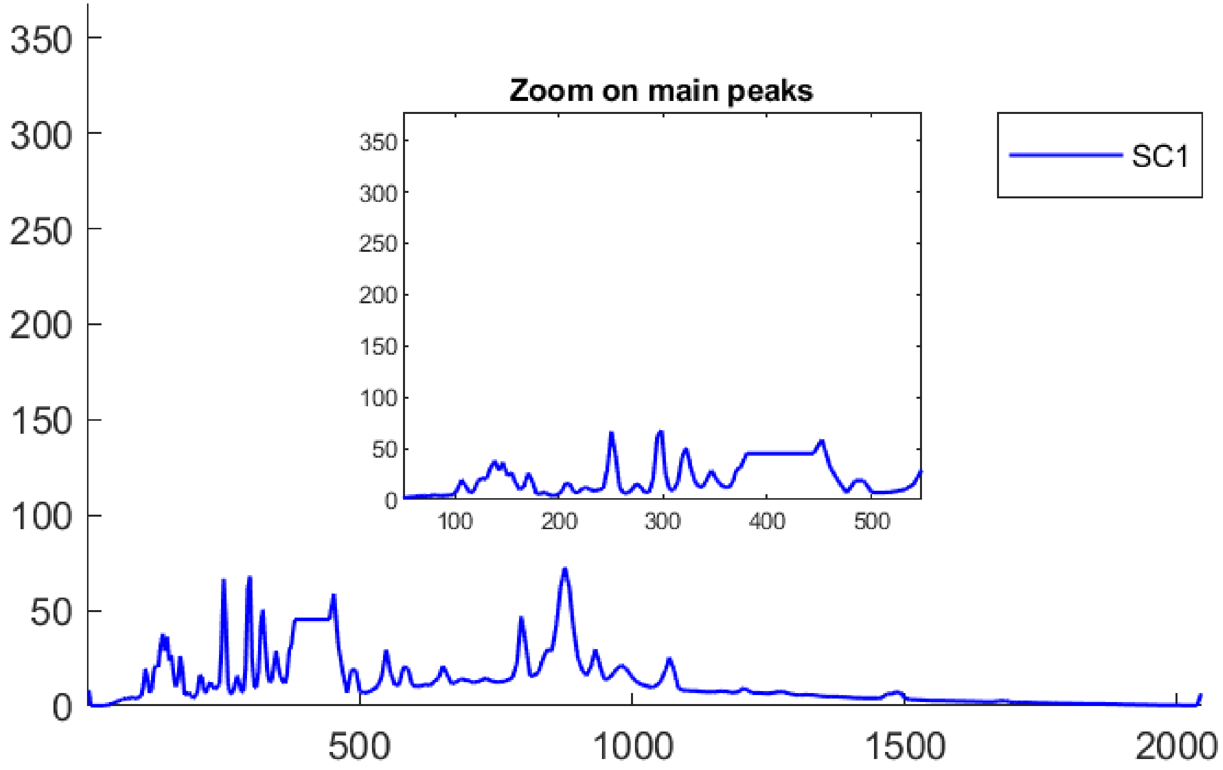
## Subclasses of class 14



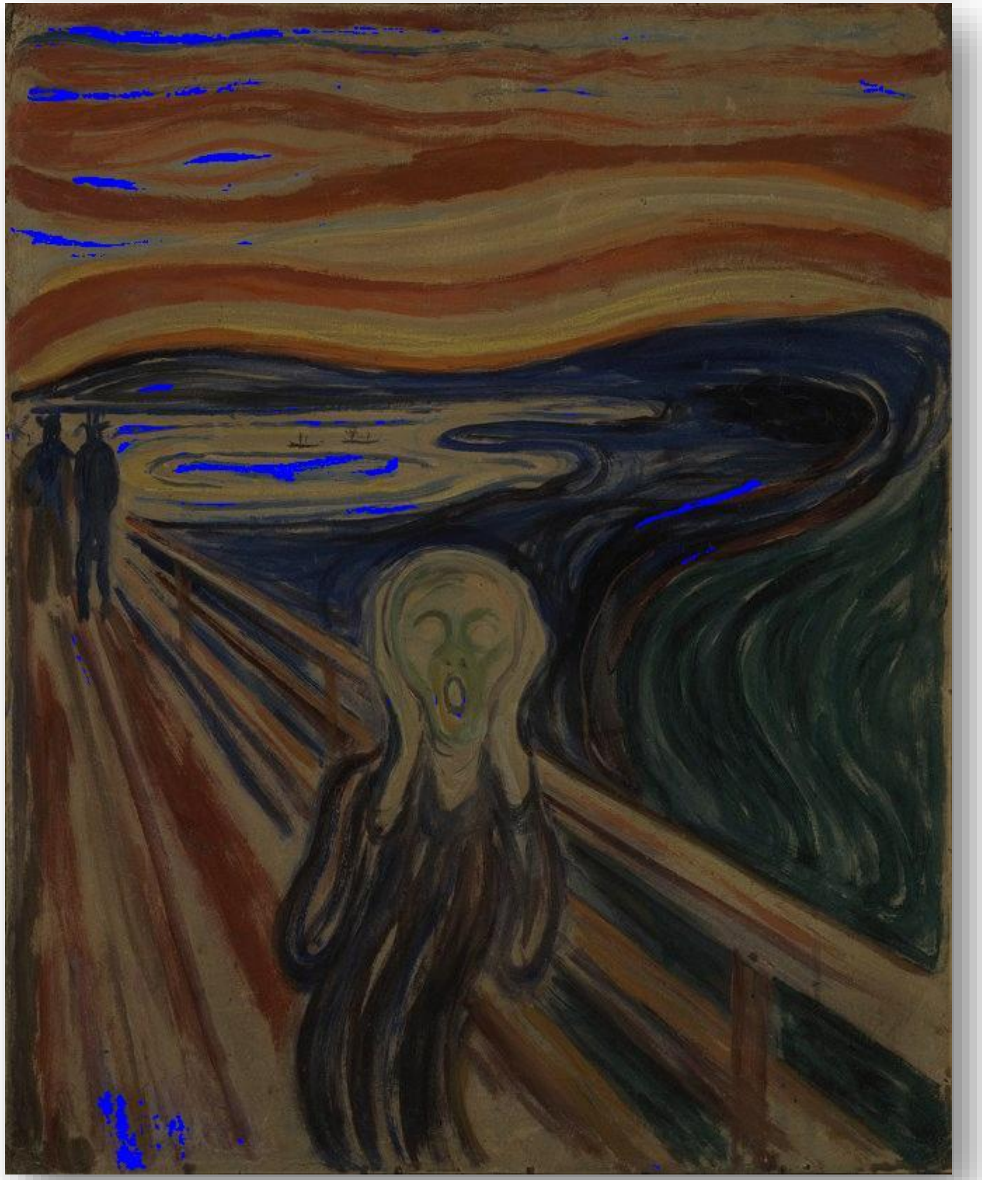



# Results - second stage

## Subclasses of class 14

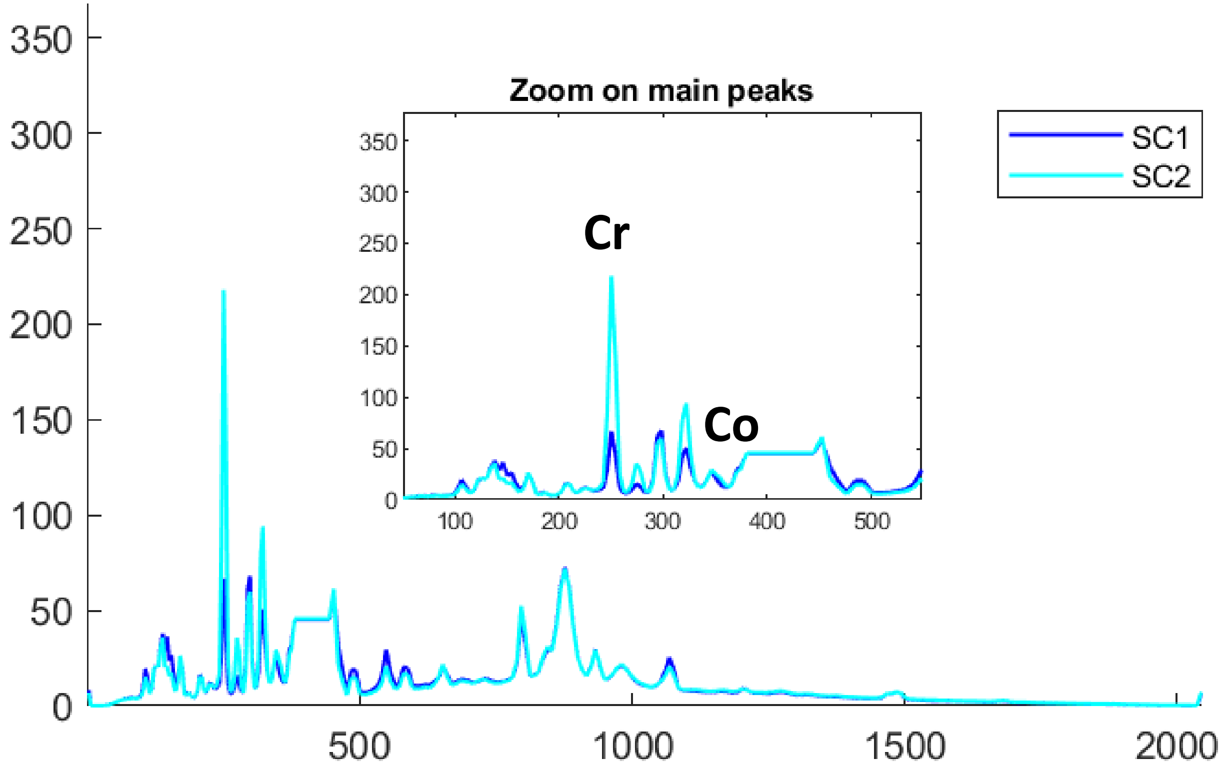


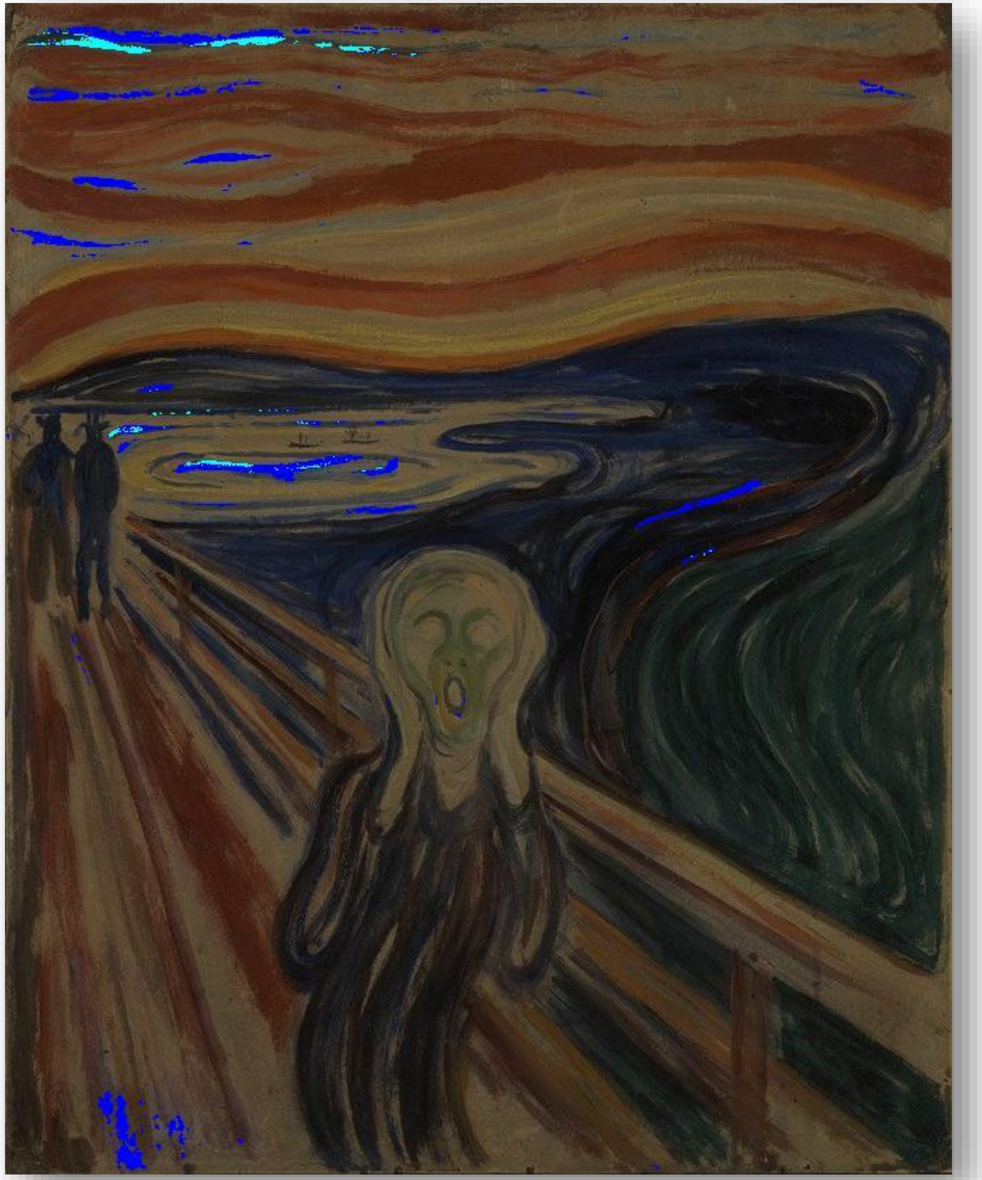

“pure” Zn



# Results - second stage

## Subclasses of class 14



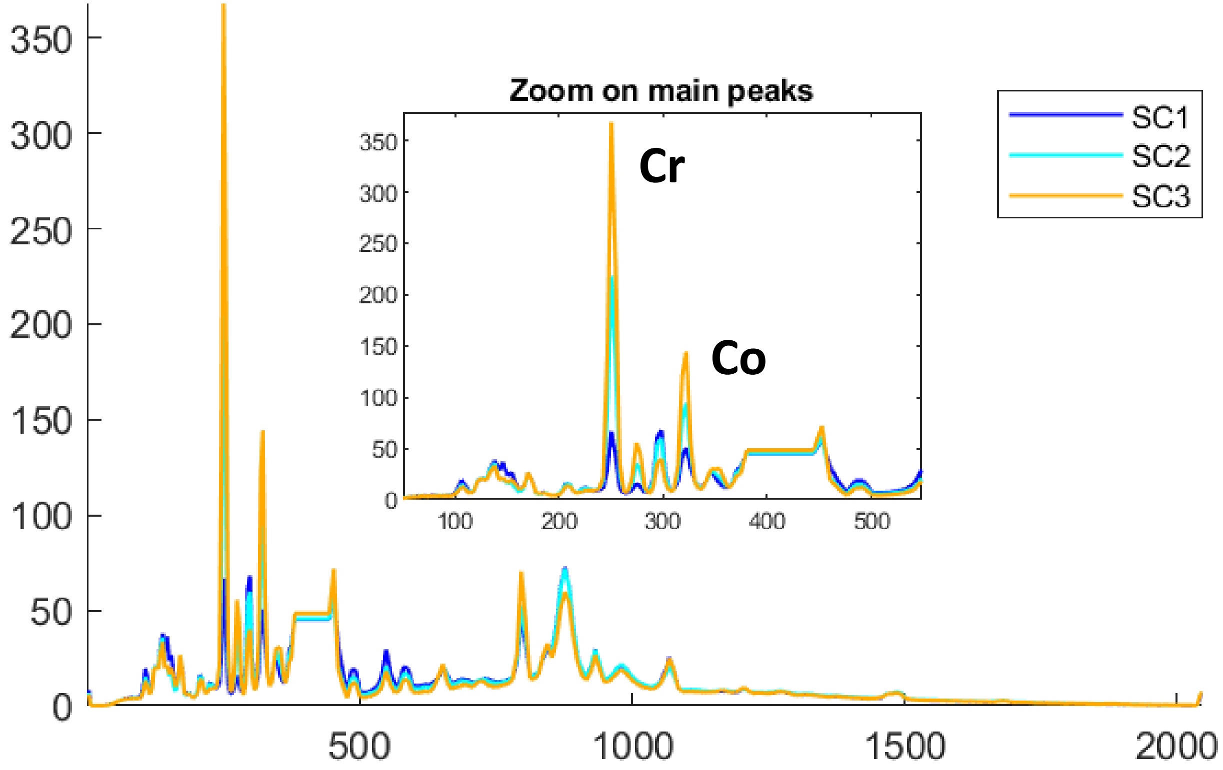
# Results - second stage

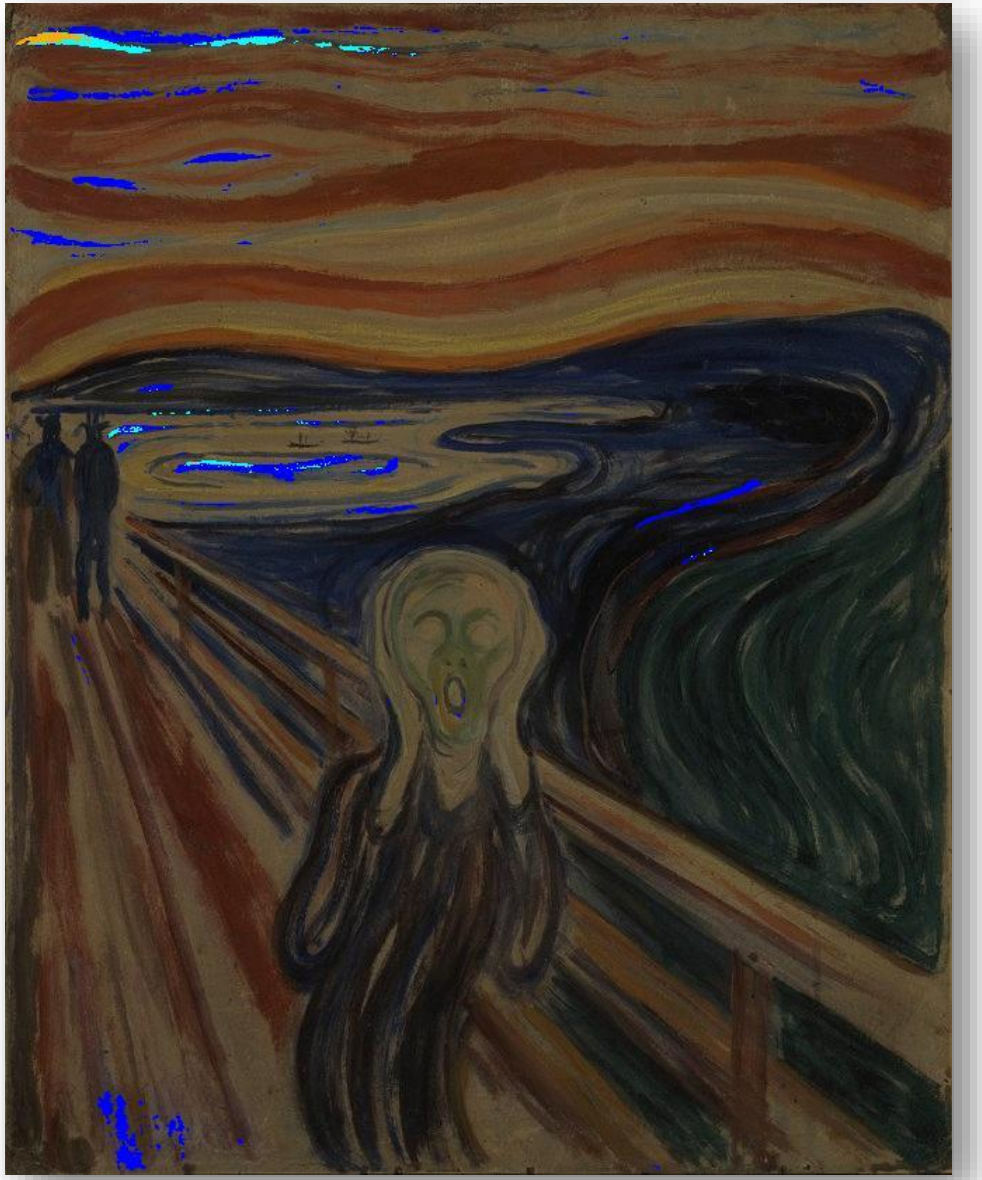
"pure" Zn

Zn - Cr - Co

Zn - Cr - Co +

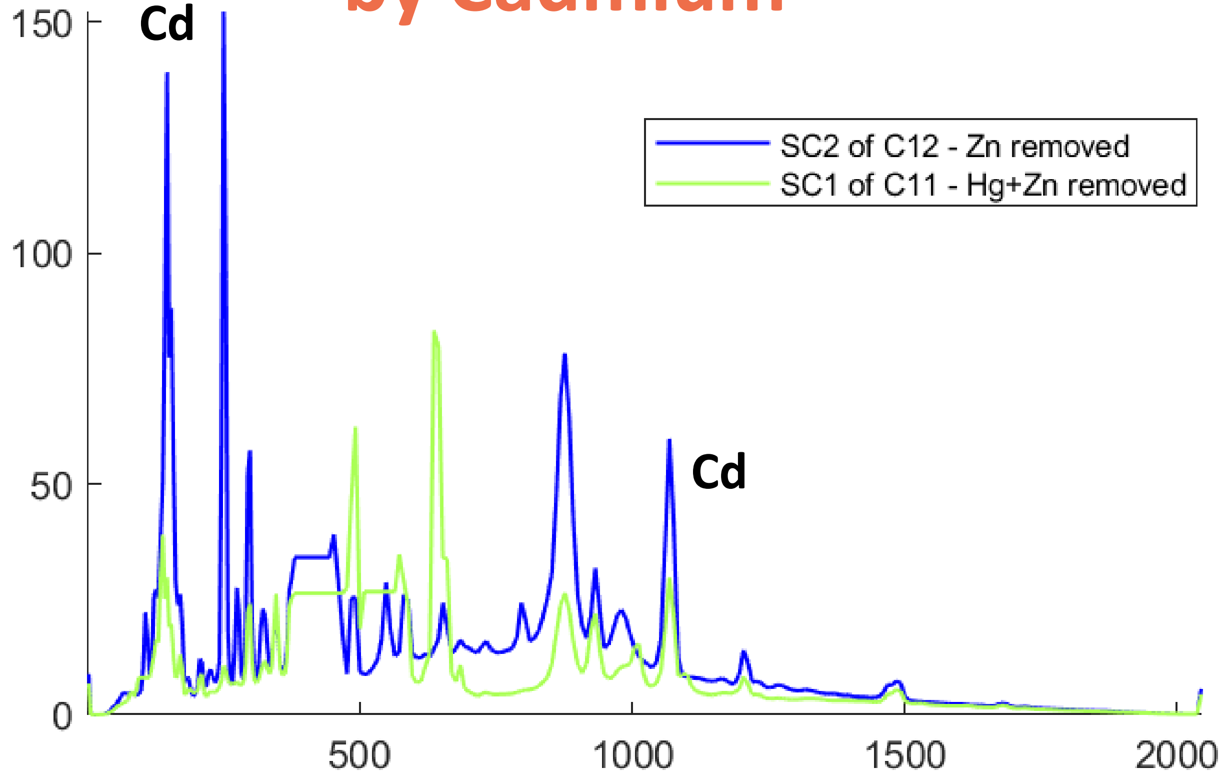
## Subclasses of class 14

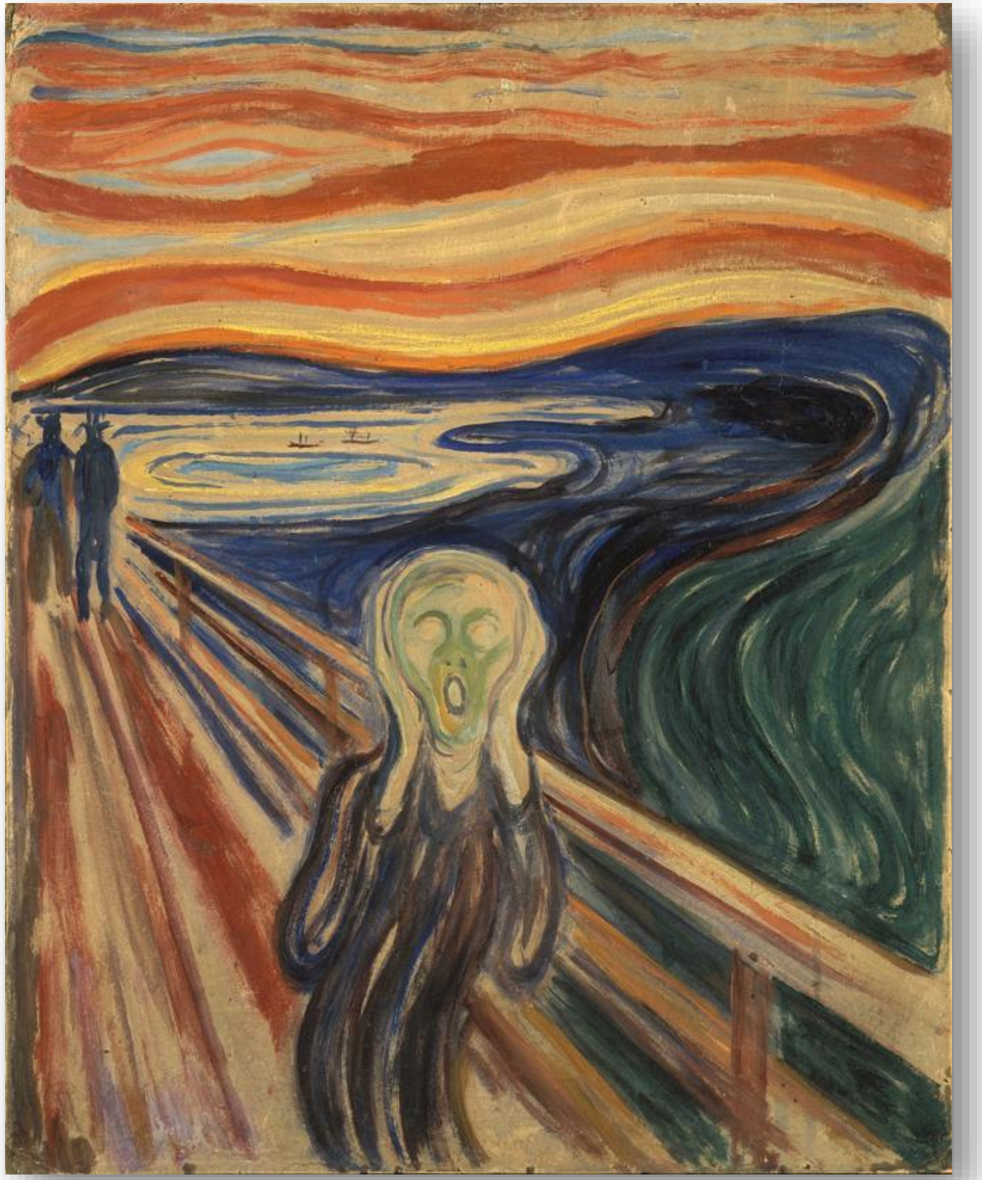


# Results - second stage

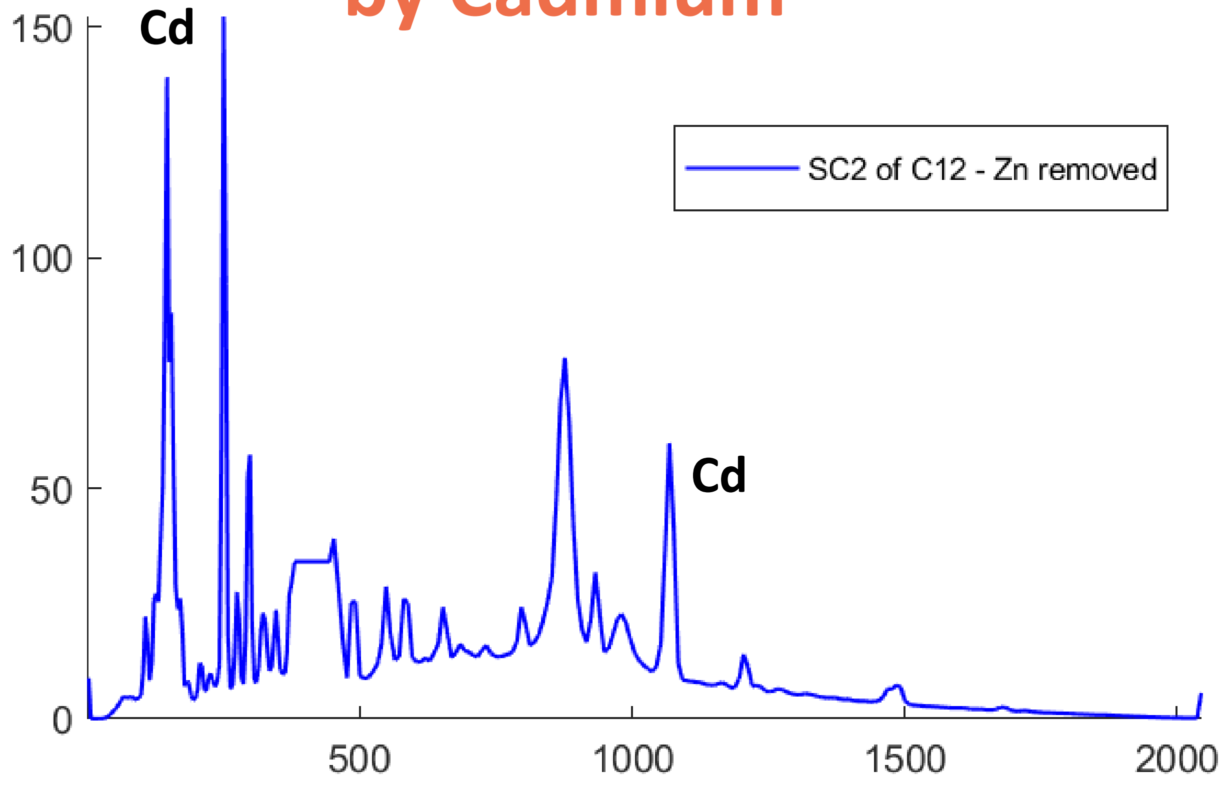
## Subclasses weighted by Cadmium



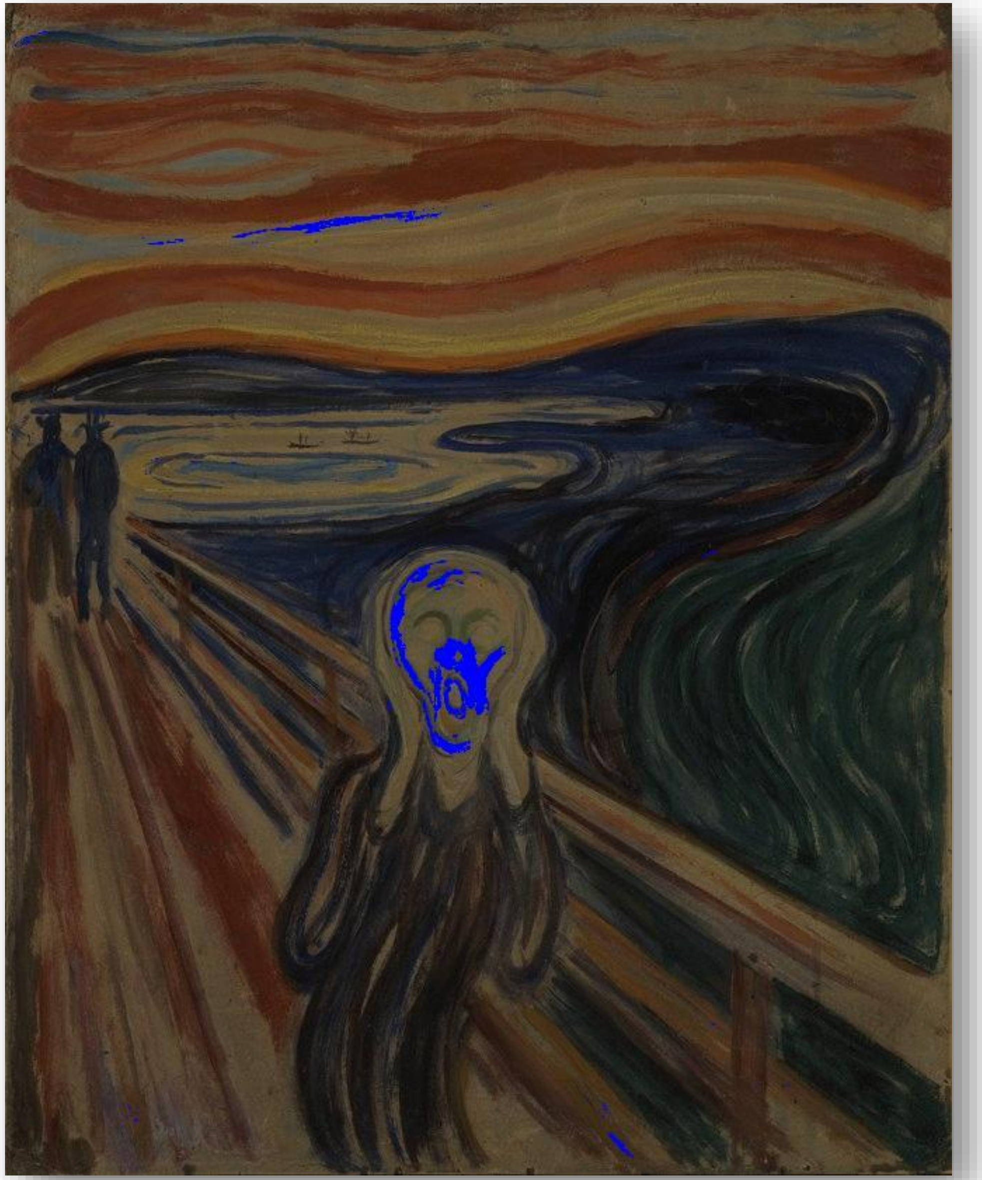



# Results - second stage

## Subclasses weighted by Cadmium




Zn - Cd

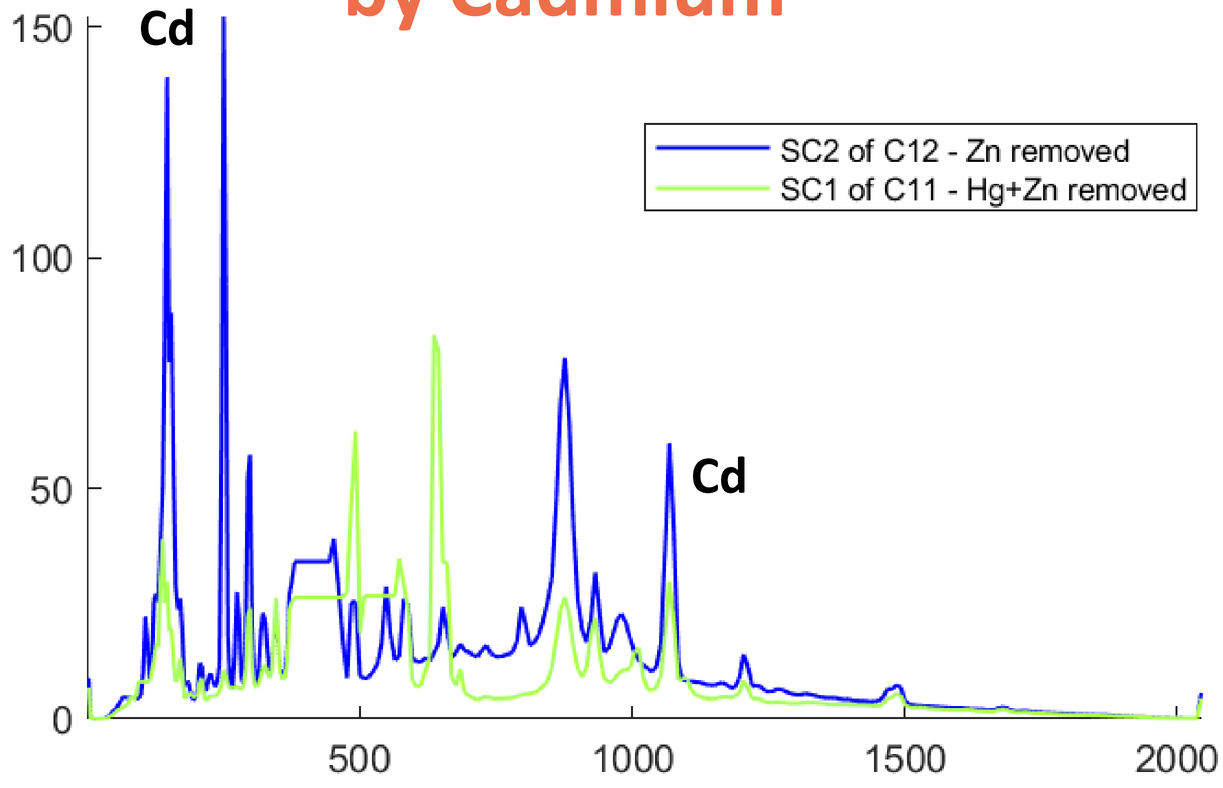


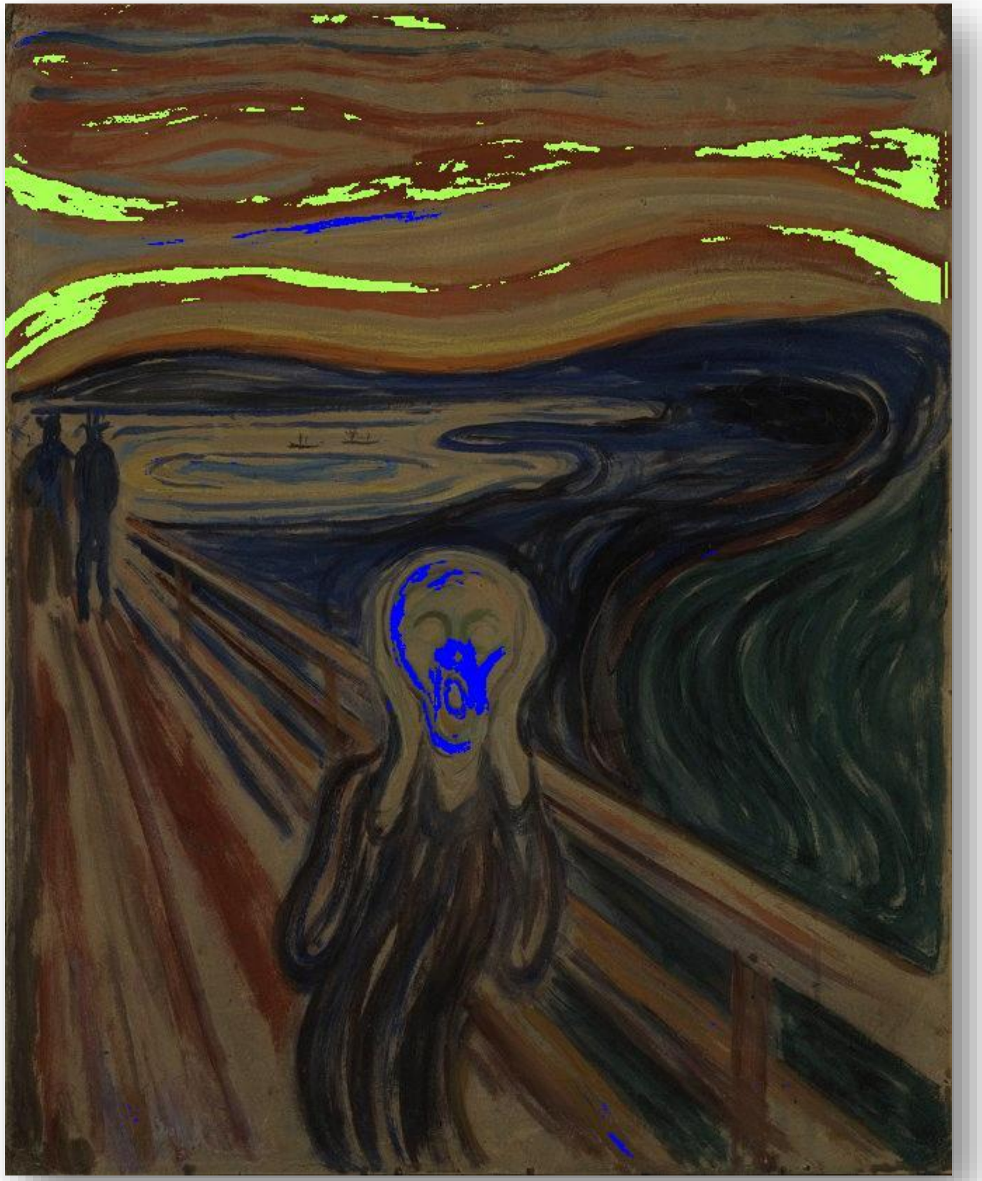
# Results - second stage

Zn - Cd

Hg - Zn - Cd

## Subclasses weighted by Cadmium



# Conclusions

**FIRST STAGE OF FCM-BASED ANALYSIS:**

Classification of the elements' distribution into four main groups:

**Zn – Hg – Cr – low signals**

# Conclusions

**FIRST STAGE OF FCM-BASED ANALYSIS:**

Classification of the elements’ distribution into four main groups:

**Zn – Hg – Cr – low signals**

**SECOND STAGE OF FCM-BASED ANALYSIS:**

Detection of combination and distribution of pigments used to create peculiar shades

# Conclusions

**FIRST STAGE OF FCM-BASED ANALYSIS:**

Classification of the elements' distribution into four main groups:

**Zn – Hg – Cr – low signals**

**SECOND STAGE OF FCM-BASED ANALYSIS:**

Detection of combination and distribution of pigments used to create peculiar shades

**ADVANTAGES:**

- Highlighting of important details
- Clarification of paint mixtures and of relations among main pigments
- Correlation between main and minor elements

# Future perspectives

**AUTOMATIZATION** of grouping operation

- For each class $C_i$, identify $p_i$ position of the main peak;
- Let $p_j, j = 1, \dots, K$, be the distinct positions identified; include in the group $G_j$ all classes whose main peak falls at position $p_j$.

# Future perspectives

**AUTOMATIZATION** of grouping operation

- For each class $C_i$, identify $p_i$ position of the main peak;
- Let $p_j, j = 1, \dots, K$, be the distinct positions identified; include in the group $G_j$ all classes whose main peak falls at position $p_j$.

**CLASS MERGING** at FIRST STAGE:

→ More robustness w.r.t. the initial choice of the number of clusters

# Future perspectives

**AUTOMATIZATION** of grouping operation

- For each class $C_i$, identify $p_i$ position of the main peak;
- Let $p_j, j = 1, \ldots, K$, be the distinct positions identified; include in the group $G_j$ all classes whose main peak falls at position $p_j$.

**CLASS MERGING** at FIRST STAGE:

→ More robustness w.r.t. the initial choice of the number of clusters

**FURTHER REFINEMENT**

- More appropriate similarity metrics or nonlinear operators
- Employing a soft clustering approach → more sensitivity
  → increased capability of emphasizing relationship between non-predominant elements

# Thank you
# for your attention!