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Anthropisation of the Rhône River during the last century: case of main pollutants measured in sediment cores

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ARCHEORHONE project (2017-2018)
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Context and objectives

The 20th century has been marked by the development of many industries such as chemical, textile, watchmaking, nuclear and mechanical fields along the Rhône River. This rapid industrial rise led to a lack of information on the environmental consequences caused by these facilities in the Rhône catchment area.

The amount and the toxicity of industrial substances led to a societal concern during the last century and the list of contaminants covered by this concern has gradually increased. First, with the monitoring of radionuclides at the downstream of NPP, then, with the implementation of the Water Framework Directive in 2000, some thirty priority substances including trace metals added to this list, supplemented in 2012 by new substances such as pharmaceuticals and pesticides.

The objectives of the ArchéoRhône project was to reconstruct the history and the origin of the river's contamination levels for three major families of contaminants: radionuclides, persistent organic pollutants (POP) and trace metals. For this study, two sediment cores were collected at the entrance and the outlet of the catchment area.


1)- Core sampling

- One sediment core was collected in a side channel of the Rhône River and located upstream of all industry and nuclear facilities of the Rhône River catchment and all main tributaries.
- A second sediment core was collected in a "casier Girardon" (groynes linked to each other by a submerged dike) near Arles, located downstream of the last nuclear facility (CEA Marcoule). The coring site is disconnected from the Rhône River since the 1960s, excepted during floods.

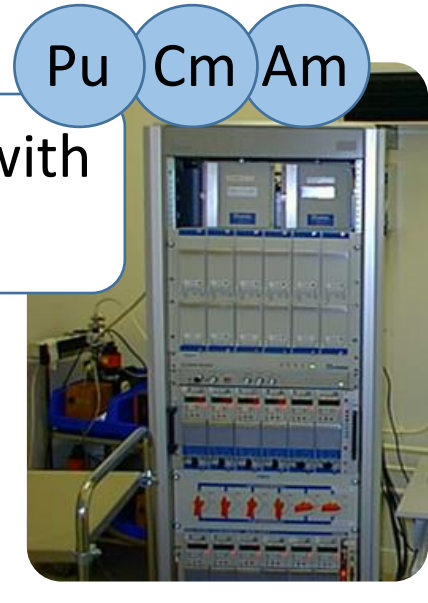
2)- Dating of sediment cores

- The dating of the core collected in the upper Rhône River was realized with the ¹³⁷Cs and ²¹⁰Pb_{xs} method (Pennington, et al, 1973 ; Appleby et al., 1993).
- Given that the peak of ¹³⁷Cs from the Chernobyl fallout was hidden by releases from the CEA Marcoule (spent fuel reprocessing facility), the dating of the second sediment core was realized using ¹³⁷Cs released by the CEA Marcoule, combining transport and deposition equations (Abril, 2018) and comparing modeled ¹³⁷Cs deposited with ¹³⁷Cs measured thanks to a spectral analysis.

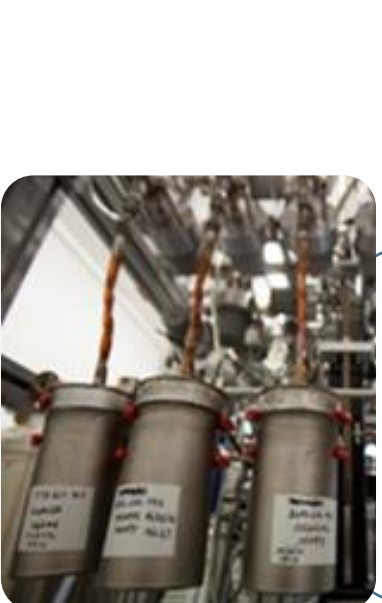
3)- Analysis of radionuclides (RN)



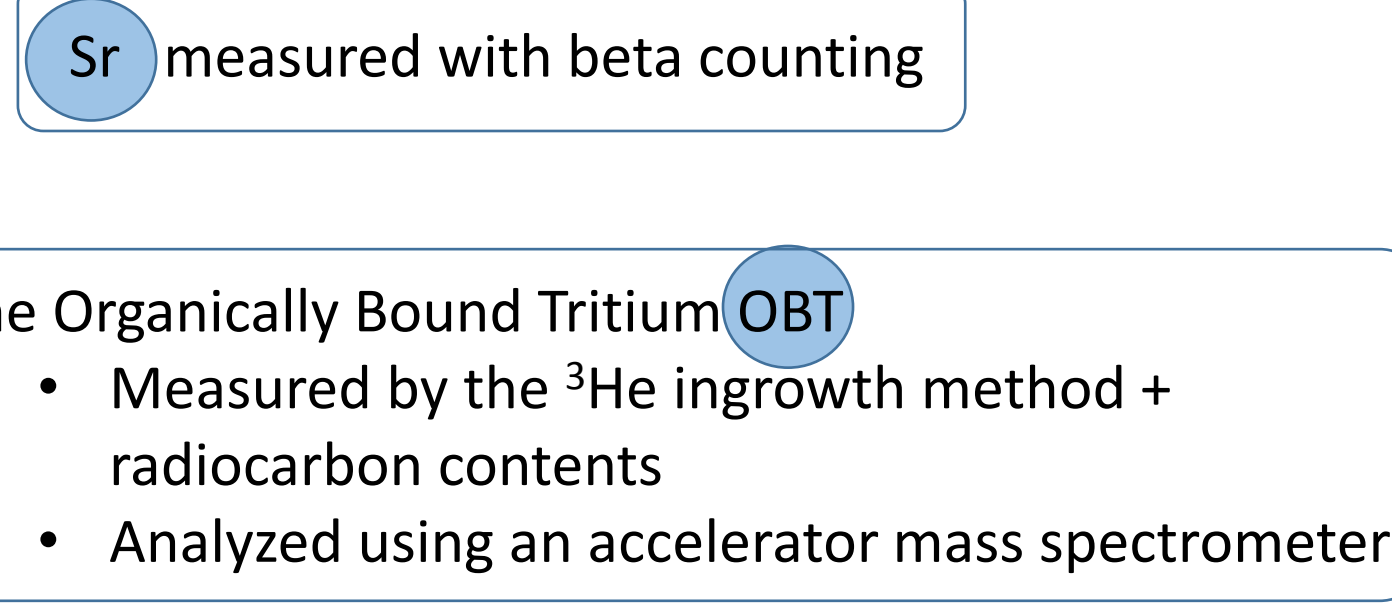
The main part of radionuclides was measured by gamma spectrometer



Actinids quantified with alpha spectrometer



Sr measured with beta counting



The Organically Bound Tritium (OBT)

- Measured by the ³He ingrowth method + radiocarbon contents
- Analyzed using an accelerator mass spectrometer

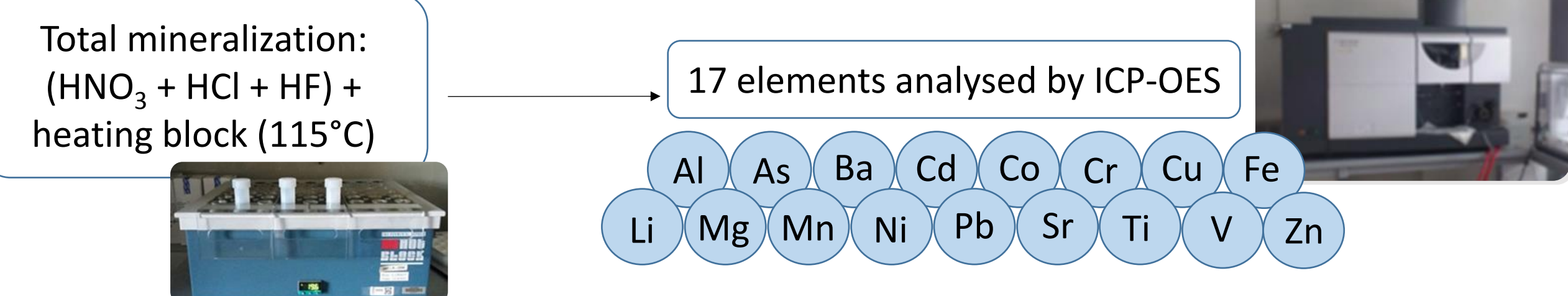
Material and method



4)- Analysis of persistent organic pollutants

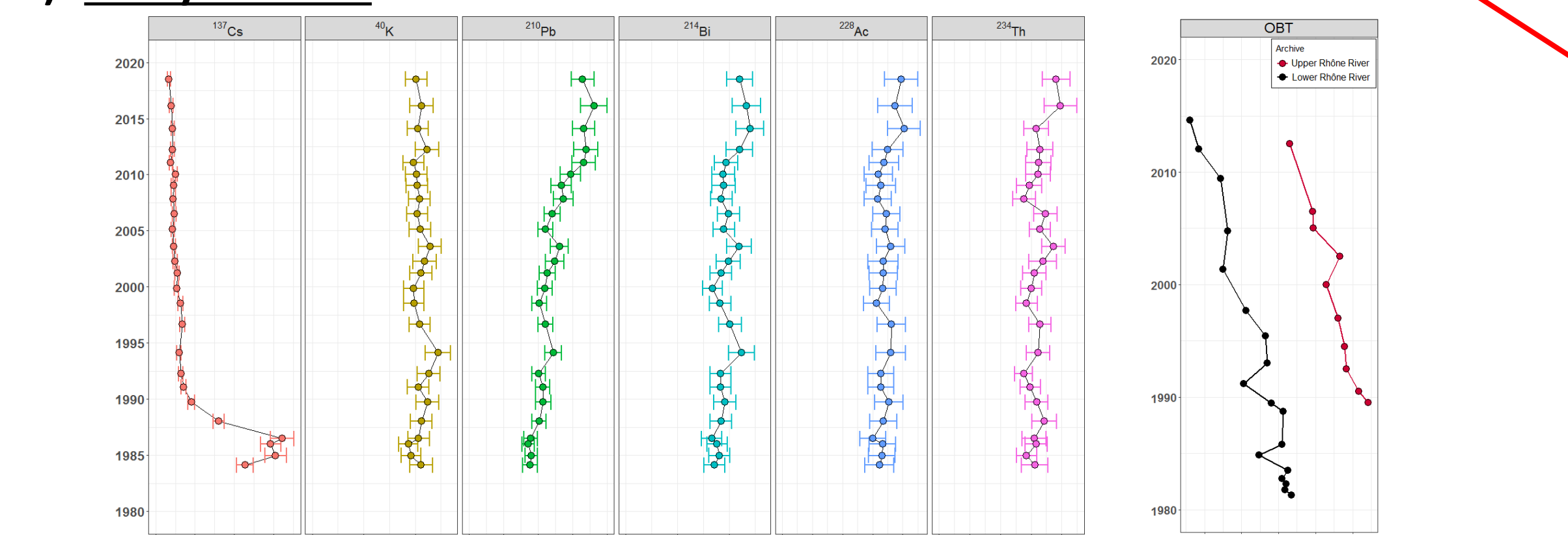


5)- Analysis of trace and major elements

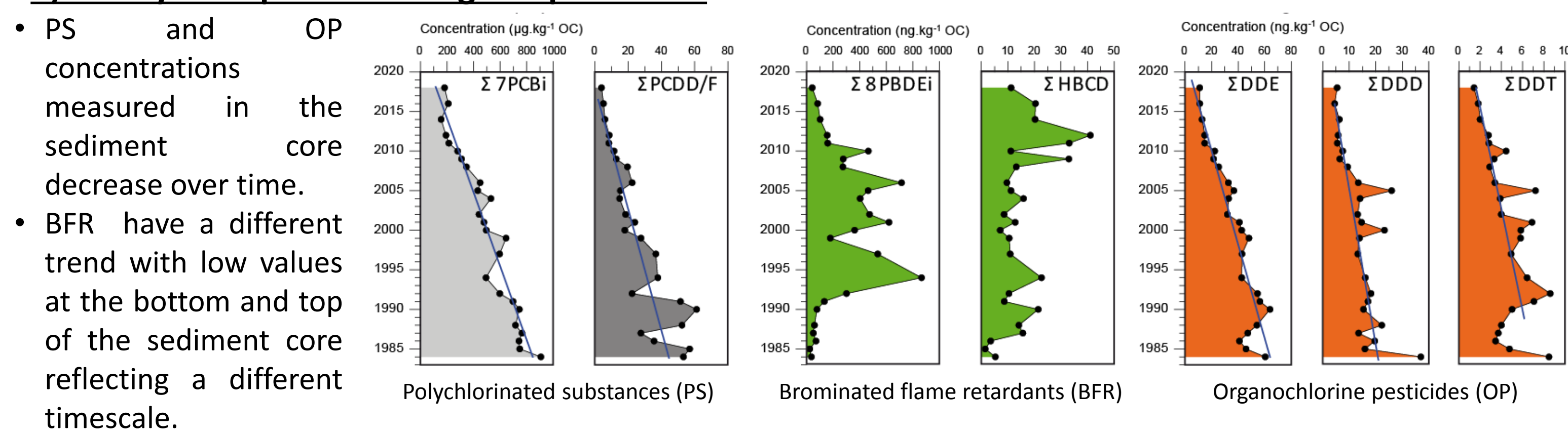


Results

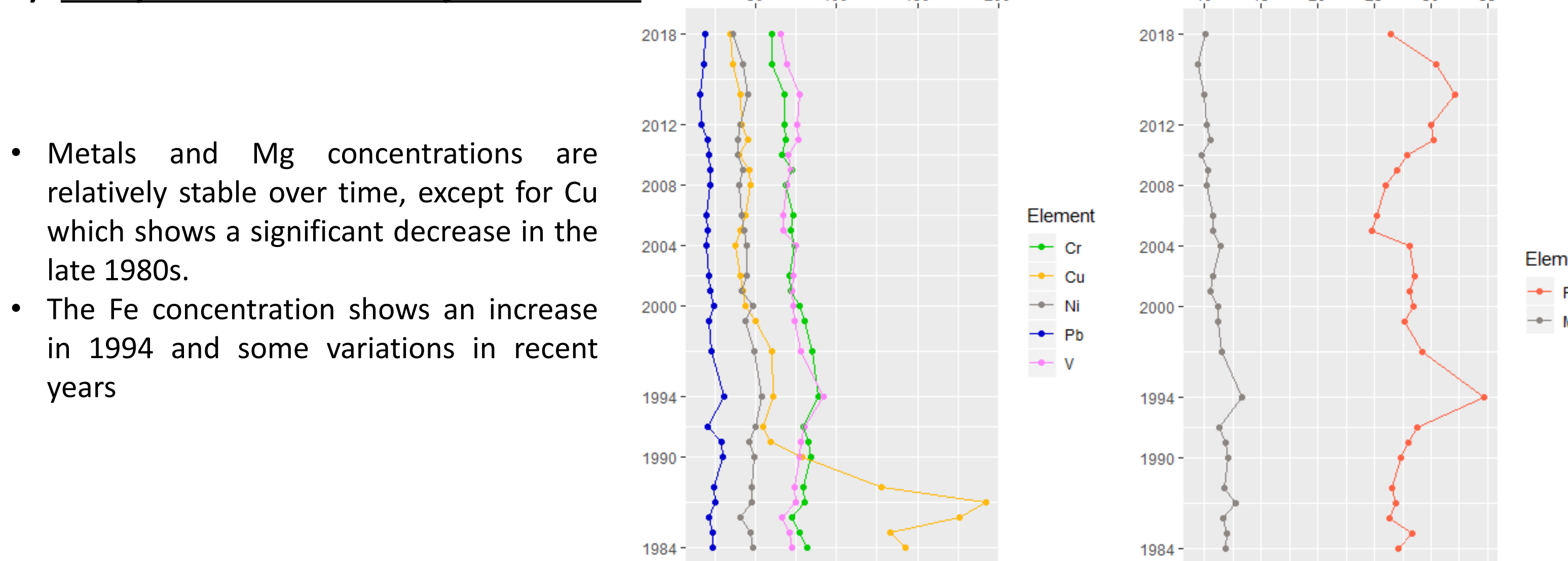
1)- Analysis of RN

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- The peak of cesium corresponds to the fallout from Chernobyl accident in 1986.
 - Concentrations of natural radionuclides are almost constant over time excepted for descendants of the U/Th chain like ²¹⁴Bi and ²³⁴Th.
 - High concentration of OBT were measured in the upper Rhône River and compared with the lower Rhône River concentrations. Those were 1000 time higher for the upstream than for the downstream. The tritium came from residues of luminescent paints used in the watchmaking industries from the 1960s to the 2000s in France and Switzerland.

2)- Analysis of persistent organic pollutants

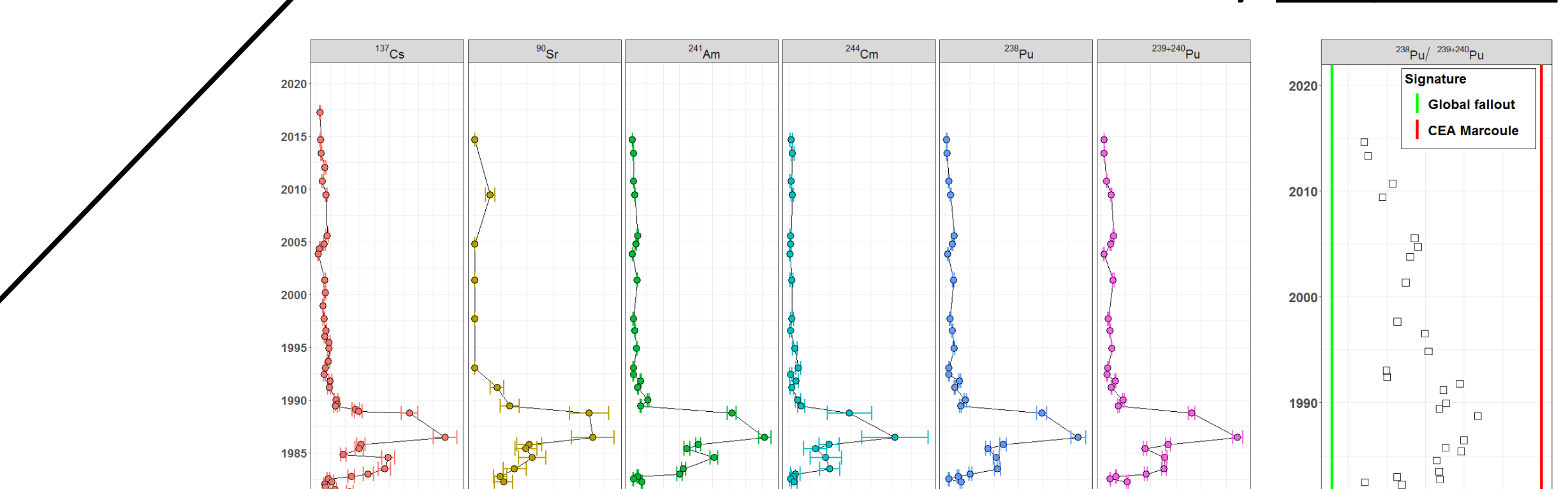
- PS and OP concentrations measured in the sediment core decrease over time.
 - BFR have a different trend with low values at the bottom and top of the sediment core reflecting a different timescale.
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3)- Analysis of trace and major elements

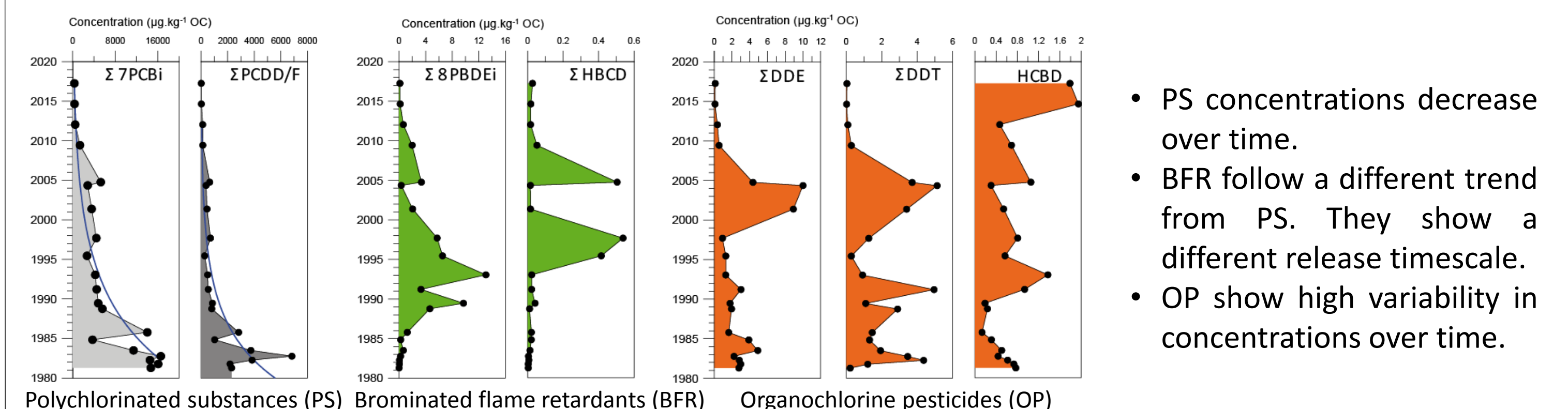
- Metals and Mg concentrations are relatively stable over time, except for Cu which shows a significant decrease in the late 1980s.
 - The Fe concentration shows an increase in 1994 and some variations in recent years
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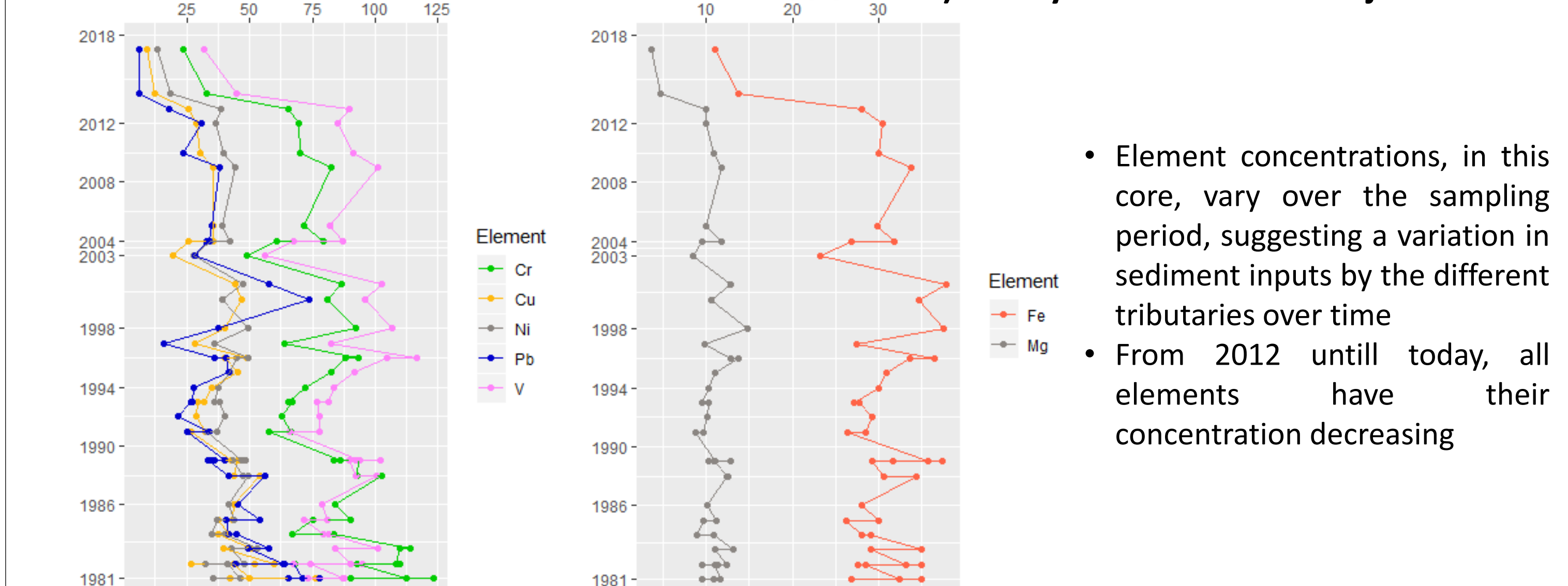
1)- Analysis of RN

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- Concentrations of artificial radionuclides are higher before the 1990s. Since 1991 the CEA Marcoule set up a new station for the treatment of liquid effluents leading to the observed decrease of concentrations..
 - Pu ratio explains the plutonium contribution from the CEA Marcoule or/and the global fallout. The contribution of the Marcoule is higher than the watershed (i.e. global fallout) contribution before the 1990s. Nowadays this contribution is decreasing showing the resilience of the Rhône River.

2)- Analysis of persistent organic pollutants

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- PS concentrations decrease over time.
 - BFR follow a different trend from PS. They show a different release timescale.
 - OP show high variability in concentrations over time.

3)- Analysis of trace and major elements

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- Element concentrations, in this core, vary over the sampling period, suggesting a variation in sediment inputs by the different tributaries over time
 - From 2012 until today, all elements have their concentration decreasing

Conclusion

- Sediment cores allowed us to quantify the amount of main pollutants transiting through the Rhône River during the last decades.
- Since the 1990s, a significant decrease of artificial RN was observed as a result of the set up of a new station for the treatment of liquid effluents of the spent fuel reprocessing facility of the CEA Marcoule which was the main source term of artificial RN during decades until the 1990s. Natural RN show relatively steady concentrations overtime in both sediment cores.
- Metals concentrations, except Cu, don't show any variation for the upstream core, unlike the concentrations measured in the downstream core. These variations in concentrations are probably due to more or less significant contributions from the different tributaries.
- Significant concentrations of OBT were measured in both sediment cores, especially in the upstream. Watchmaking industry led to a chronic contamination of the entire river, and even more near the watchmaking workshops.
- PS concentrations decreased progressively over time since the 1980s mainly in the downstream core. The increase of BFR from the 1990s to the 2000s suggests a different timescale. The OP trajectory is variable and seems more complex. Concentrations of the majority of substances studied were higher before the 1990s (RN, PS, trace elements) or between the 1990s and the 2000s (BFR). Since this period the Rhône River started a resiliency phase.