Reading Herculaneum Material using X-rays

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Disclaimer

- I googled some pictures for illustration, apologies if they lack proper references or credits
- Whatever is extracted from my own work is unpublished
  - Not because I don't believe in peer-review publications, but because there is more work involved and results are never quite good enough yet to warrant working on publications rather than improvements
- Wherever you're missing a plot or information, feel free to get in touch and I'll try to provide them later
- Also - this is **always ill prepared** and I will **keep talking forever**: **Intervene**, tell me the time and to get on, ask questions

Outline

- Introduction to the topic
- A bit about the Light Sources I went to
- Existing measurements
- Why am I here
- The "latest and greatest"
- Summary

Why the Volcano? (Introduction)

- In 79AD Mt Vesuvius erupted and over a few days destroyed much of the surrounding area and life within
- During archaeological digs in the area in 1750, a Villa was uncovered and a library discovered within
  - After dumping the first few tens or hundreds of charred scrolls into the sea, people realised
  - If you want to see a life size model, travel to the Getty Villa in LA
- All in all, some 2000 charred scrolls were recovered
- Few have been read to this day

Why not read them then?

Stereotypical old book as found in your standard Oxford College Library

Why not read them then?

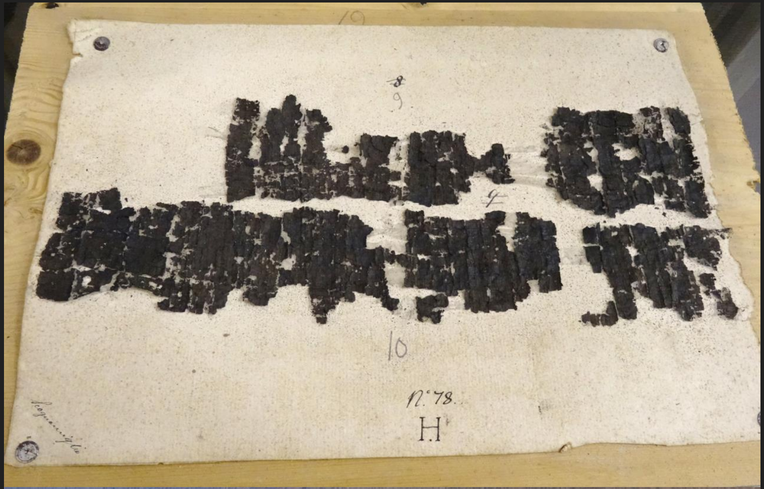
Stereotypical scroll from Herculaneum (not in standard OX Coll. Lib)

Why not read them then?

- Subjects have been carbonised through being covered in hot ash (heat, no oxygen)
- Otherwise perfectly preserved
- Early attempts at unrolling succeeded,

  somewhat

- Outer layers of scroll can be adhered to a flexible film and peeled off
- Inner Layers (most writing) tend to break of in lumps…



Interlude 1 - Light Sources (Involved)

Light sources involved with Herculaneum

- ESRF, Grenoble, France
- Diamond Light Source, Didcot, United Kingdom
- Canadian Light Source, Saskatoon, Canada (Phantoms)

Where I have worked on Herculaneum material

- Diamond Light Source:
  - Beamline I12:
    - High Energy: 53-150keV
    - High Flux: ~2e11 photons/mm^2/s
  - Bealine B16:
    - Low Energy: 4-45keV (unfocussed)
    - Low Flux: 1e9 photons/mm^2/s
- Canadian Light Source:
  - Beamline BMIT:
    - High Energy: 28-140keV
    - High Flux: 5e10 photons/mm^2/s

Applying for Beamtime

- Pay-for access available - at DLS that is somewhere around 5k (reasonable currency) per 8h (ballpark, with no claim for accuracy)
- All light sources have a peer review process for beamtime requests submitted
- So far as I can tell successful applications depend on an upfront discussion with a beamline scientist, to make sure what one is proposing can actually work at the beamline
  - The more worked out and thus viable a proposal seems, the less likely it is to end on the "hopeless" heap
  - There does seem to be an influence on whether the project has something unique/peculiar that tends to get people excited:
    - Making XRD measurements of yet another makeup …
    - Making 2000 year old burned scrolls readable for the first time…

Some general thoughts on light sources

- Support varies not by light source, but by beamline and is crucial, even when you bring everything yourself
  - Just running the beamline in an ideal mode makes a big difference
- Recording efficiencies matter: Having a brilliant light source and massive photons flux does not mean brilliant stats through your sample!

- ESRF has just undergone works, Diamond undergoing works now in various places
  - More photons yet, at more high intensity beamlines yet

Existing Herculaneum Measurements

Existing Measurements

- Early measurements on material composition in a single letter snippet from a Parisian scroll showed lead contamination in the ink, which was not visible elsewhere on the papyrus
- CT scans of these scrolls did not show any ink visible in absorption, but ink was distinguishable on surfaces

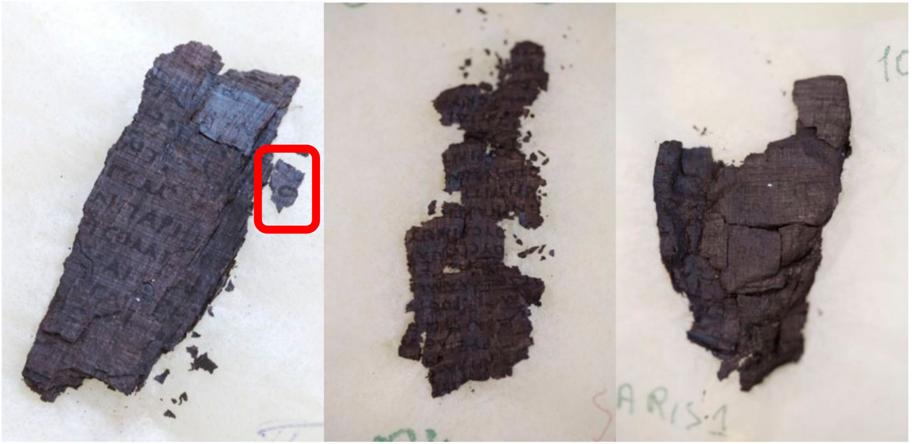


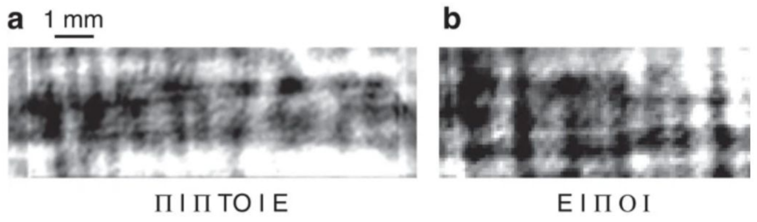
Figure 3: Three Herculaneum fragments on loan to the University of Kentucky in 2008: PHerc.Paris. 2, fr. 96, 126, and 101.

Existing Measurements

Mocella, V., Brun, E., Ferrero, C. et al. Revealing letters in rolled Herculaneum papyri by X-ray phase-contrast imaging. Nat Commun 6, 5895 (2015). https://doi.org/10.1038/ncomms6895

- Approached imaging by means of phase contrast, ballpark 60keV - Lead electron density should deliver a large change in refractive index -> phase propagation differs
- I find this very hard to believe, as the subjects themselves have various sizes of structures embedded within them,
  and seeing the minutia of lead contamination in a massive structure made from compressed fibres …?
- I am not an x-ray scientist :-(
- They also did some surface XRF
- Note that this group is good with Nature

Figure 1: Two words read on a hidden layer of the fragment *PHerc.Paris. 1* fr. 101.



(**a**) The sequence of Greek capital letters ΠΙΠΤΟΙΕ (*pi-iota-pi-tau-omicron-iota-epsilon*); (**b**) The letter sequence of the next line, ΕΙΠΟΙ (*epsilon-iota-pi-omicron-iota*).

Interlude 2 - Why am I here

Curiosity

- A physicist walks into google offices
- Got roped into a discussion with Brent Seales (University of Kentucky) about ways to approach Herculaneum
    - Got the full brief on what they had done to that day
    - Came up with differential imaging and (thanks to a colleague) XRF CT measurements as a way forward
- Waited 3 years to find all the right people in all the right places to get going
    - Basically no funding, i.e. bits were loaned, time spent was "hobby", beamtime was granted courtesy of the beamline (Diamond I12, thanks Michael)

"New" Measurements

Novel stuff (Machine Learning)

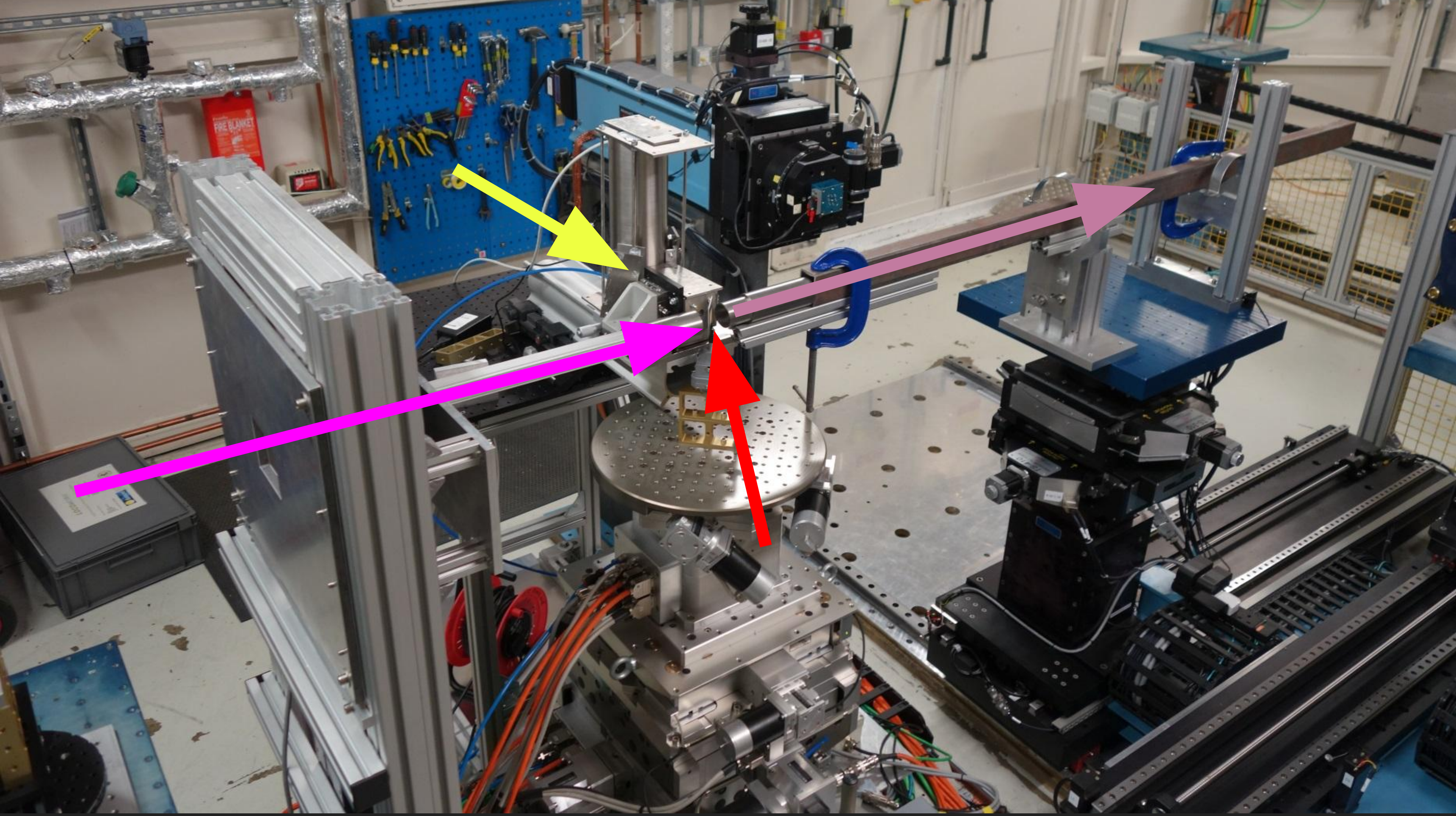
Parker CS, Parsons S, Bandy J, Chapman C, Coppens F, Seales WB (2019) From invisibility to readability: Recovering the ink of Herculaneum. PLoS ONE 14(5): e0215775. https://doi.org/10.1371/journal.pone.0215775

Previous and new CT scans approached with machine learning, using visible layer scans to inform truth information and train an evil AI to find ink on hidden layers - They now have funding to support an open contest to do this better:

https://scrollprize.org/

XRF CT

- Came up with the idea in 2013, got first measurements in 2016:
    - Object is illuminated with x-rays above the k-edge (Lead: 88.0045keV)
    - Energy/Laterally resolved sensor is placed at an angle to the primary beam, observing the object through a pin hole
    - Magically, images of material composition shall appear


FIRE BLANKET

XRF CT

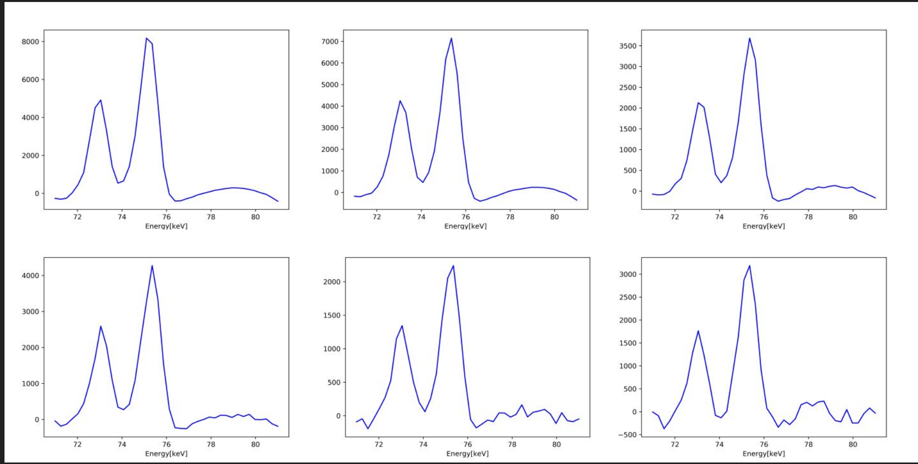
- Came up with the idea in 2013, got first measurements in 2016:
  - Object is illuminated above k-edge (Lead: 88.0045keV)
  - Energy/Laterally resolved sensor is placed at an angle to the primary beam, observing the object
  - Magically, images of material composition shall appear
- First test was pretty much made from prototype detectors and scraps (including random shaped plates of tungsten shielding)
- Second attempt was made in 2017 with 5+ days of beamtime, tungsten flight tubes and yet not enough to get a decent setup working
  - Energy calibration prevents pixel summation
  - Largely recorded at one observation angle and injection Energy

Results from flat imaging in XRF (k-lines)

Made our own leaded ink, different concentration levels from X down to X/1000

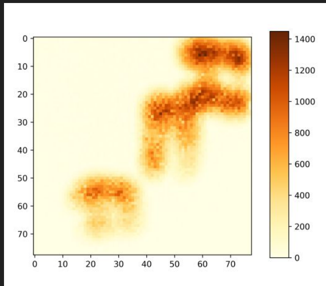
X/300 is similar to our target, most imaging measurements made at X/10

Recording times up to hours

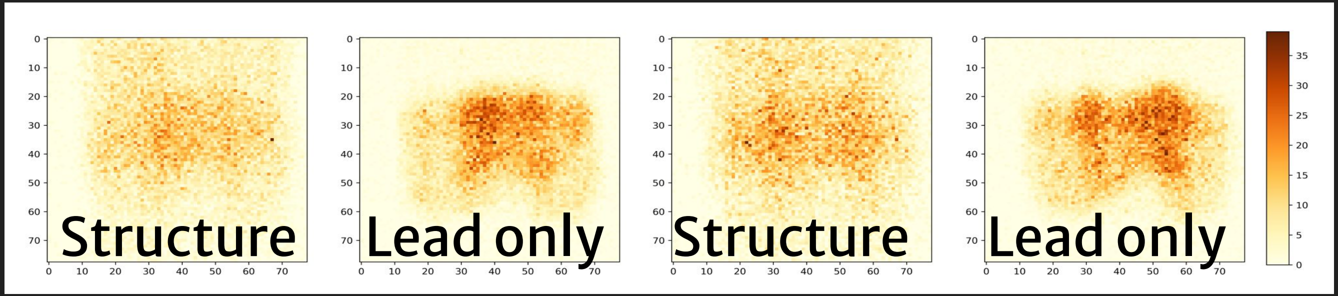


Concentration	Counts s/pixel
Alpha	10.5734
Beta	1.0102
Beta/3	0.1990
Gamma	0.0782
Gamma/3	0.0177
Delta	0.0115

Results from flat imaging in XRF (k-lines)

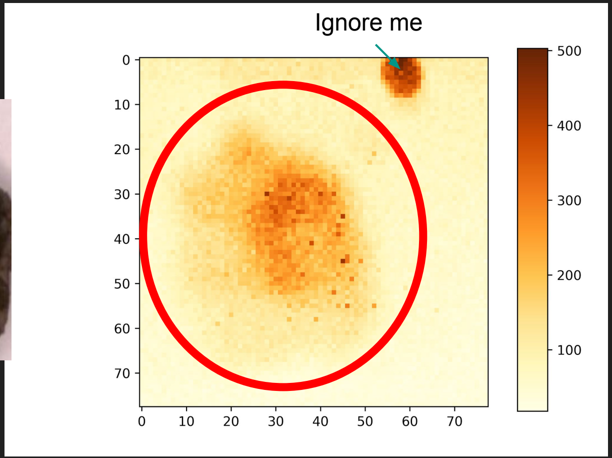


- Multilayer scans show no measurable degradation in lower layers
- First CT scans with a rolled up sample:
  - Structure and ink can be observed differently, selecting either compton scatter or fluorescence energys (with some background magic)
  - Stats are horrendous and this was at high concentrations of lead

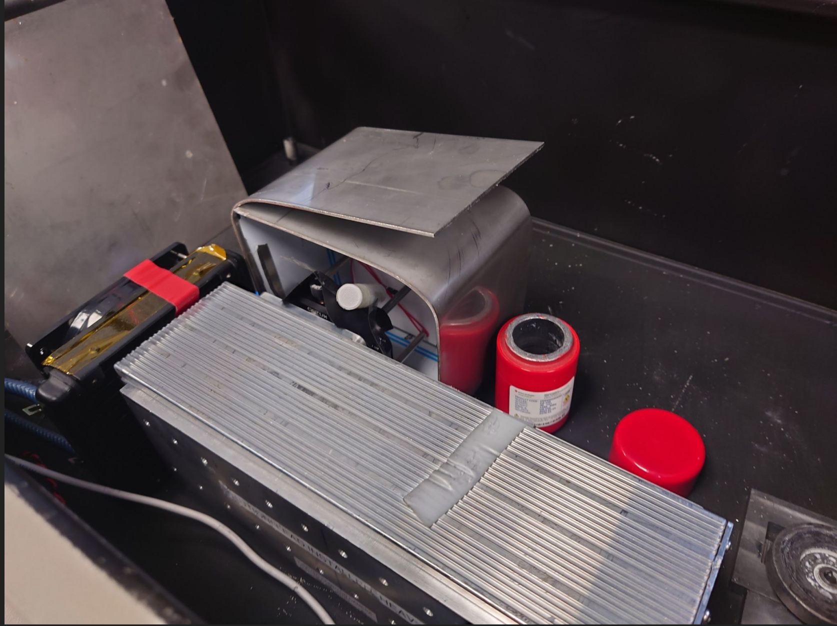


Results from flat imaging in XRF (k-lines)

- We can see k-line fluroescence in the ink!
  - Just…
  - After 8h for one flat image
- Problem clearly not solved
- Issues:
  - Efficiency of the setup is crap (1e-12 - ish)
  - Injection energy and compton scattering overlaps signal
  - Detector noise leaves long tails even to higher energy
  - Beam Energy less well defined than hoped for
- Bid with the Arts and Humanities Council covered purchase of 109Cd source
  - Highest emission energy is 88.035keV, 30.5 eV above lead k-line
  - Observation under large scattering angles should set the Compton peak will below k-lines



Bench top setup



- Tried it earlier this year:
- No, it's not sophisticated, but not an issue
- Fluorescence is well visible in L-lines
  - Down to about X/30, somewhat X/100
  - Definitely not X/300
  - Many hours recording time
- Pretty much nothing in K-lines
  - Dominant emission from 109Cd is 22 and 26keV, 88keV is only 2%
  - Higher activities need a custom approach, E&Z will only deliver standard sources to 40mCi, as the self absorption of the lower energies makes higher activities inefficient to produce - most other customers (handheld XRF scanners for example) care about the lower energies

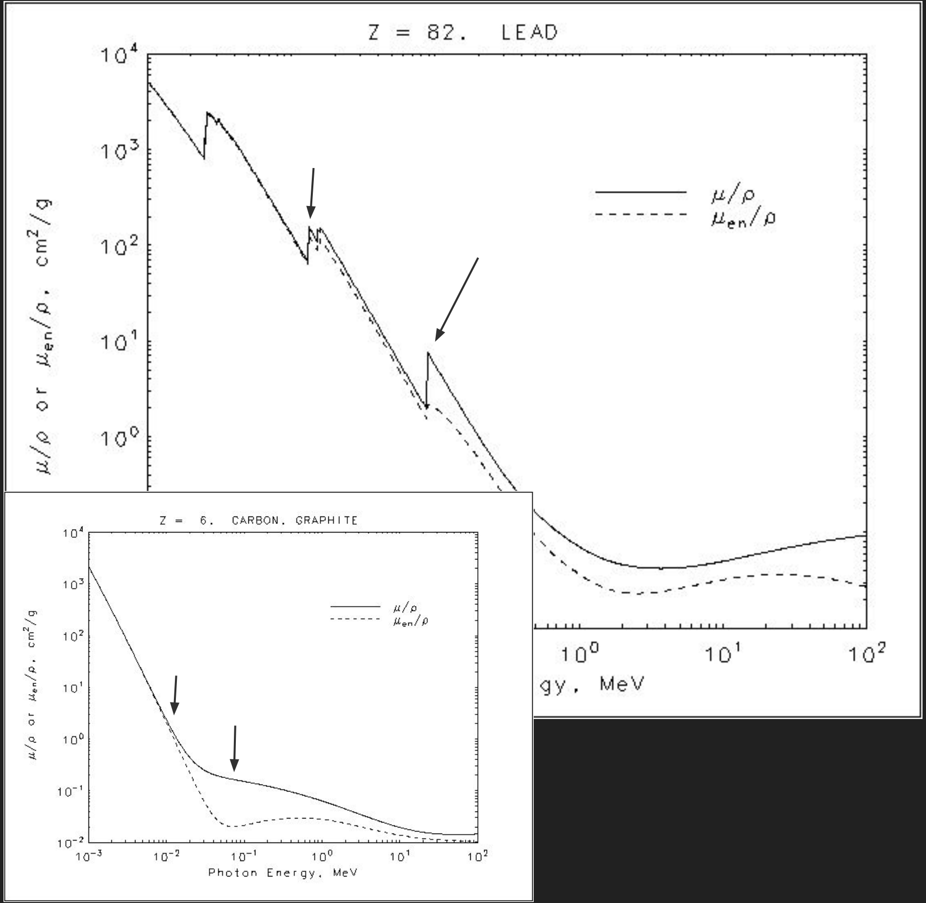
:-(

Didn't you mention "Differential" imaging?

- Given the enthusiasm I had about XRF, I never spent much time on it
  - Till last year
- An ATLAS Colleague was spending time at CLS and roped me in for a discussion about my problem
  - Had 5 Beamline Scientists in a video call, all keen to help figure this out
  - Proposed K-line edge imaging and got beamtime for August last year
- Went away and spoke to Diamond colleagues, got rapid access granted on B16 for L-edge imaging

Differential/Edge imaging?

- Run 2 CT scans, one below and one above an absorption edge of your favourite element (lead)
- Difference between these should only show the different behaviour in absorption across the energy change
- Lead K-line: Factor 7 up
    - Carbon constant(ish)
- Lead L-line: Factor 2.5 up
    - Carbon dropping (but assume constant)



Does it work?

No!

Well, not quite yet

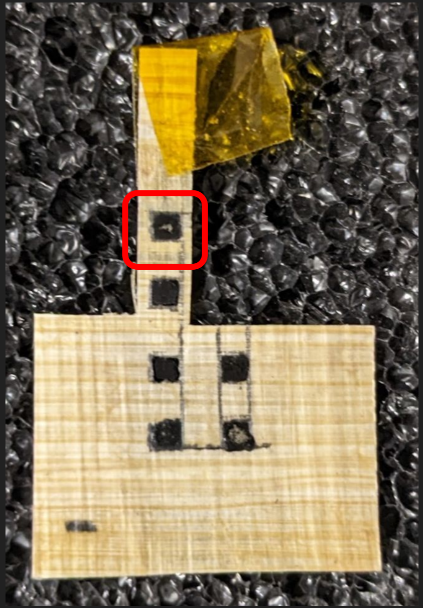
CLS: K-lines

Little hope for actual sensitivity due to very low absorption rate, but this was 2 days with amazing efficiency already, can easily see contrast for X/30

Diamond: L-Lines

Find the pattern (you're a factor of 3 above Herculaneum concentration, but the image is part of a [fast-ish] CT scan)

- Large amounts of phase contrast
- Massive inefficiency in data recording
- Sample wasn't firmly mounted
  - Motion is amplified in differential images
  - We improved this at CLS, knowing what we knew from Diamond
- Should go back and try better…
  - Beamtime request was rejected for lack of: Publications!



Conclusion?

- More differential imaging beamtime at CLS coming up
  - Better recording efficiency yet, some more ideas about how to improve the processing to up the stats
  - Unlikely to solve the problem
- Need to get more time at Diamond, but Diamond-II upgrade might prevent that (plus my constant lack of time to work on this)
  - Decrease detector distance
  - Enclose flat sample in a support structure
- CT reconstruction is then a given, and the scan below the corresponding edge can provide information for Brent's virtual unfolding chain (requiring structural data) to then map the differential data onto flattened subjects
  - This might **"just"** work

Conclusion

- All of the above is a hobby
  - Very little time
  - Typically 0 budget
  - Very much supported by my employer in other ways
- It got me to places like the Institute de France (Paris), the inner parts of the Bodleian Library (Oxford), the Oxyrhynchus Papyri Collection (Oxford), the Scientific Side of the Getty (LA, thanks Brent), Research sections of the Natural History Museum (London) and the Chester Beatty Library (Dublin)
- It is certainly something that enriches my life and makes me feel like I am contributing something useful/interesting to society
- I made it into the Daily Mail (not a bucket list item of mine…)

Appendix

Appendix A - Conversations with Conservators

- Unrolled objects have been glued down on soft supports - that doesn't help
- General approach in conservation is to keep the object from harm (first and foremost)
- Establishing imaging methods requires establishing boundary conditions and some inner parameters - often in conflict with conservation

Appendix B - on the topic of light sources

- Diamond people are friends
  - Reasonably easy to get a hold off (for me), which is invaluable
  - Once I had the right people, they are always trying to help!
- CLS people are Canadian (read: a-ma-zing)
  - Once you've applied successfully with a proposal, you can re-apply against the same proposal with a new workplan - makes life a lot easier if you're doing the iterative tests towards a working method!!!
- Data: Typically a few TB, anywhere between 1 and 3 so far