THE IMPORTANCE OF PARTICLE INDUCED X-RAY EMISSION (PIXE) ANALYSIS AND IMAGING TO THE SEARCH FOR LIFE ON THE OCEAN WORLDS

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Introduction:

In searching for evidence of life on Ocean Worlds, detection & quantification of the biogenic elements C,N,O,P,S is critical. Detection of the cations of the rock-forming minerals (Na, Mg, Al, Si, K, Ca, Ti, Cr, Mn, Fe) and anions such as Cl, Fl are also important in establishing context.

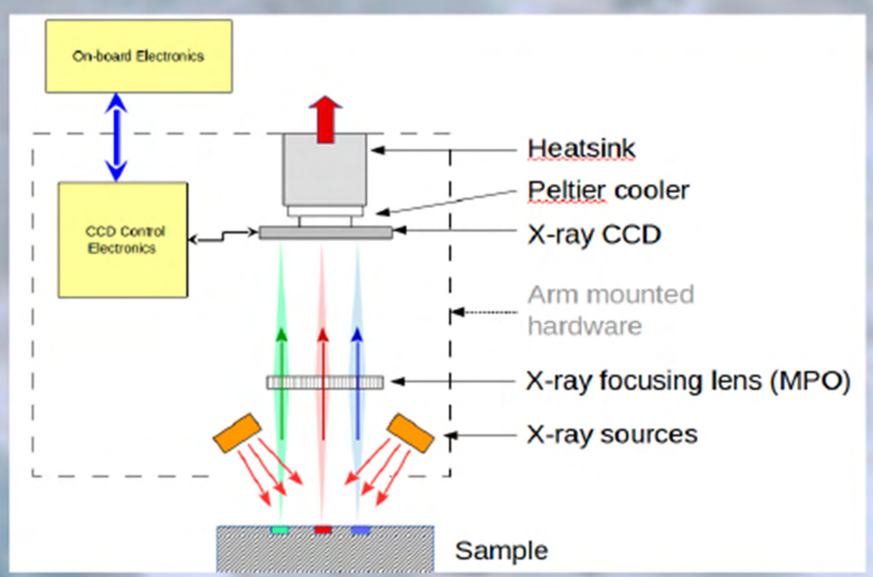
As a fluorescence source, ^{244}Cm (used in all of the Alpha-Particle X-ray Spectrometer (APXS) instruments to date [1-4]) is preferred because the α-particles at 5.8 MeV strongly fluoresce the lower atomic number elements and the γ-rays at 14 and 18 KeV adequately fluoresce the mid-range elements Ca – Mo. However, these sources are not available within NASA.

We suggest that it is strategically important for NASA to support the development of ^{244}Cm radioisotope sources for use in PIXE-based life detection and other *in situ* elemental analysis instruments.

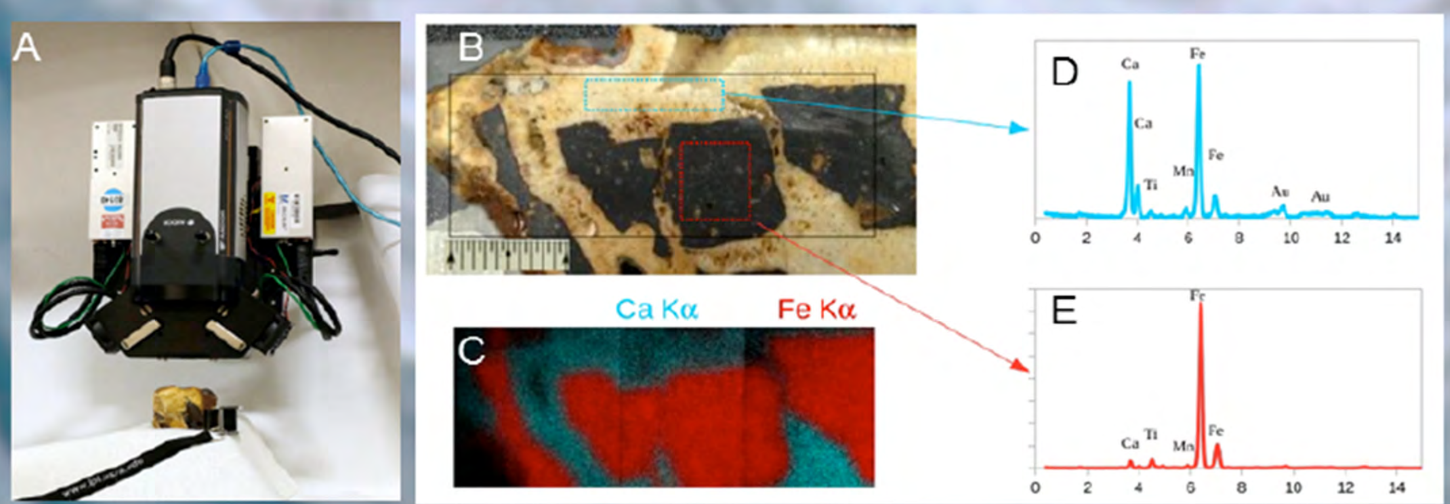
The detection and spatial co-location of the biogenic elements C,N,O,P,S on or in a mineral/ice substrate would strongly support evidence of prebiotic processes or of extinct/extant life.

MapX: a microscopic elemental imager

MapX [5], intended for the payload vault of the Europa lander, will directly image the biogenic elements C, N, O, P, S, as well as the cations of the rock-forming minerals (Na, Mg, Al, Si, K, Ca, Ti, Cr, Mn, Fe) and important anions such as Cl, Fl. The instrument provides element images with ≤ 100 μm lateral spatial resolution over a 2.5 cm X 2.5 cm area, as well as quantitative XRF spectra from Regions of Interest (ROI).



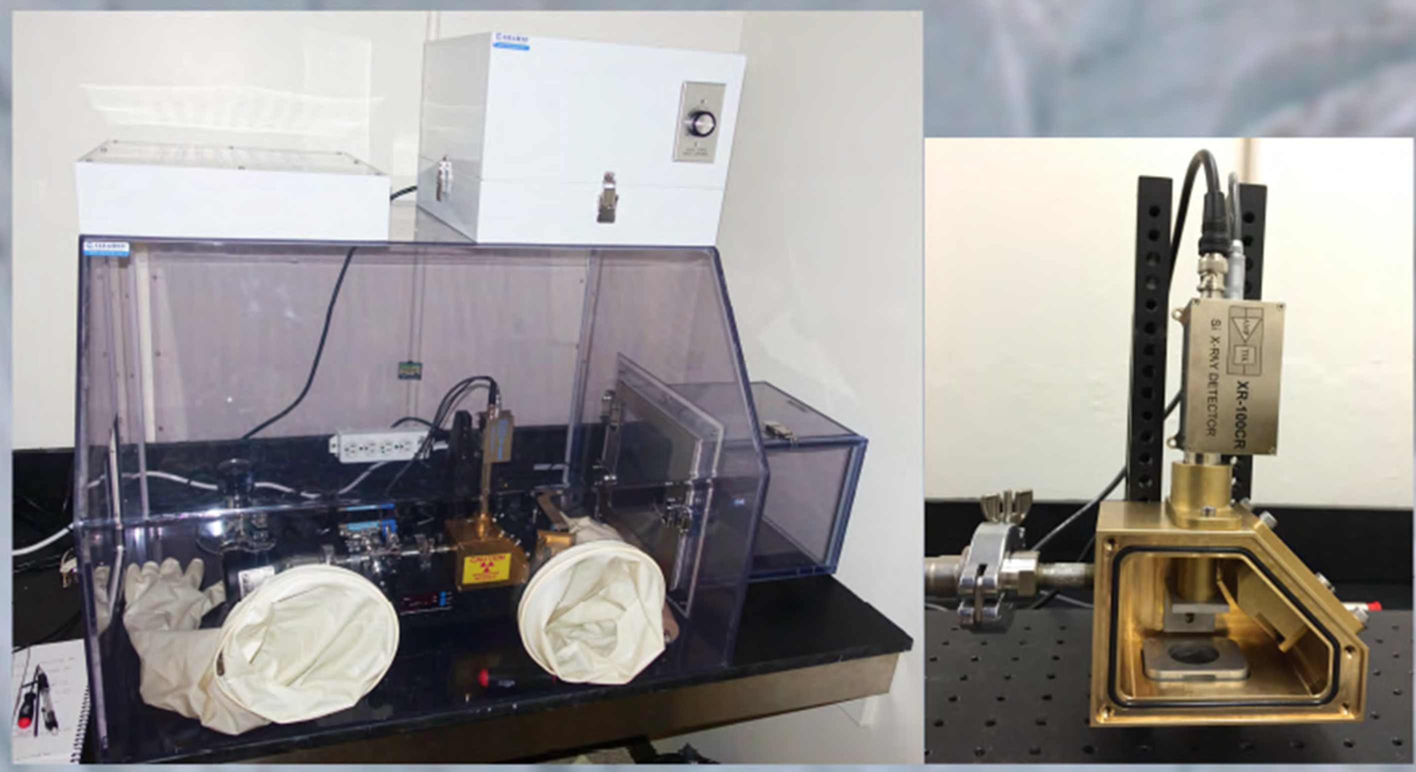
Principal components of MapX: X-ray/γ-ray/α-particle source(s) illuminate the sample. Fluoresced X-rays pass through a Multi Pore Optic (MPO) lens and are focused on a CCD detector that records the x,y positions and energies of the photons.



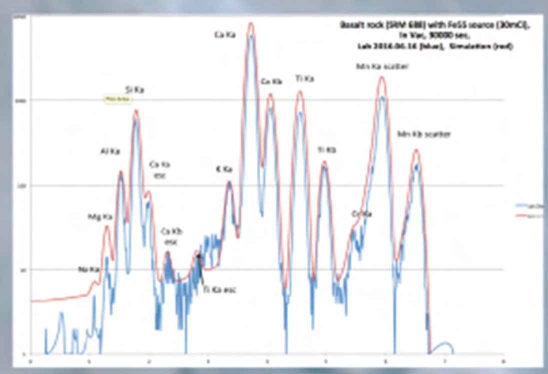
MapX prototype and example data. A: MapX-II prototype, B: Optical image of sample (scale in mm), C: Elemental image of sample (red = Fe, blue = Ca) D-E: XRF spectra from ROI.

Empirical evaluation of fluorescence from X-ray, γ-ray and α-particle sources:

An XRF test fixture was fabricated to study fluorescence from known mineral and rock samples. Spectra were collected in air or vacuum using either an X-ray tube or a radioisotope source. An X-ray tube source, 30 KeV, 100 μA, as well as several radioisotope sources (30 mCi ^{241}Am, 30 mCi ^{55}Fe, 50 μCi ^{254}Cf) were tested. Both ^{241}Am and ^{254}Cf are α-emitters; however both were found to be unsuitable as sources due to high background fluorescence from Compton scattering.



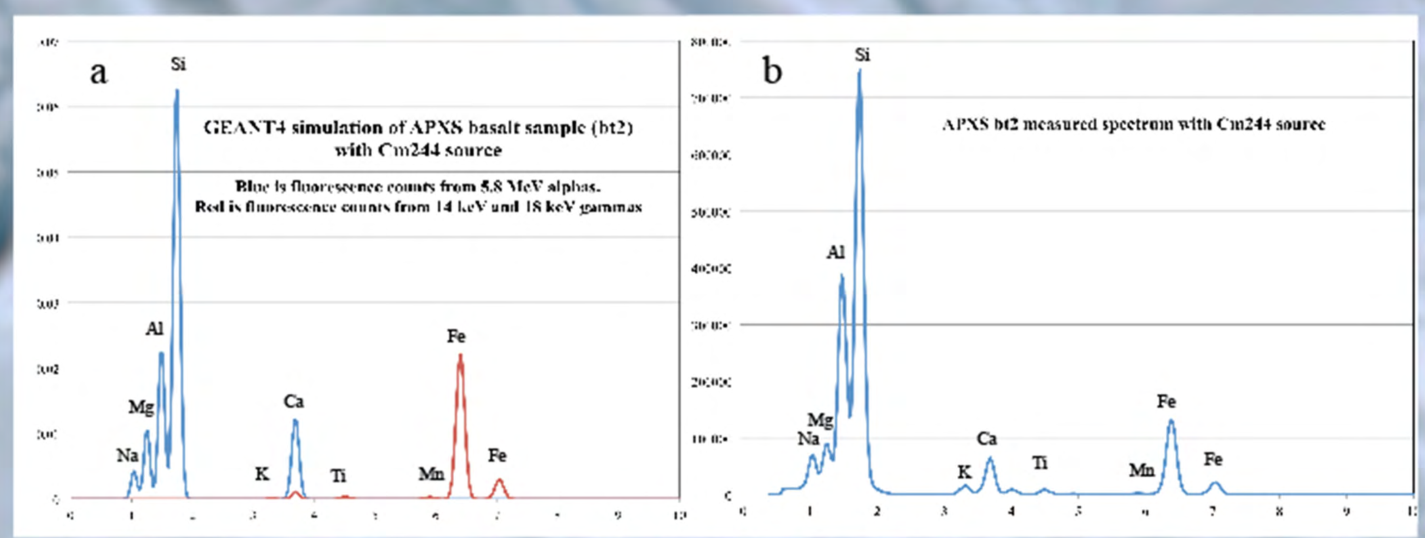
Left: Negative pressure HEPA-filtered glove box housing the XRF test fixture and pumping system. Right: XRF test fixture with radioisotope source installed.



XRF spectrum of NIST Basalt 688 using a 30mCi 55Fe γ-ray source measured in vacuum in the XRF test fixture (blue) vs. XMIMSIM [6] Monte Carlo simulation (red).

Modeling ^{244}Cm fluorescence for an imaging PIXE instrument

Because ^{244}Cm sources are not available, we are employing a combination of empirical measurements and modeling utilizing PyMCA [7], XMIMSIM [6] and GEANT4 [8] to determine source flux requirements.



Comparison of fluorescence from Basalt sample bt2 using: (a) ^{244}Cm source with 5.8 MeV α-particles, 14 and 18 KeV γ-rays modeled with GEANT4 vs. (b) published fluorescence data from the APXS instrument.

Source requirements development:

We used XMIMSIM and GEANT4 to evaluate detection and quantitation limits, defining significance level by:

$$k = N_i / \sqrt{h_B n_{ch}}$$

where N_i is # of counts above background in the peak, n_{ch} is # of MCA channels under the peak, and h_B is the height of background at the peak.

Case 1 shows results for Astrobiologically important elements relevant to rocky planets or planetesimals in two different matrices using a 30mCi ^{244}Cm (α + γ) source (first value) or 30 keV, 100 μA tube source (second value), assuming a 10^4 sec. accumulation time.

Case 2: shows the significance level k for biogenically important elements contained in water ice on an icy planetesimal (Europa), 30 mCi ^{244}Cm source, 10^4 sec.

Case 3: shows the significance level k for Biogenically important elements contained in residue from a lyophilized Europa ice sample, 30 mCi ^{244}Cm source, 10^5 sec. (zero background substrate).

Case 1: Significance Level k for Astrobiologically important elements relevant to rocky planets or planetesimals, *30 mCi ^{244}Cm source vs. Au X-ray tube source (30 KeV, 100 μA)*

		Basalt matrix		SiO_2 matrix	
Trace elements: detection @100 ppm: k >2 indicates detection at 95% level of confidence					
Element	Energy (KeV)	^{244}Cm	Au Tube	^{244}Cm	Au Tube
P Kα	2.02	1.9	10	1.9	22
S Kα	2.31	3.8	20	4.2	51
Cl Kα	2.62	3.0	35	4.2	90
K Kα	3.31	3.5	85	3.9	210
Ti Kα	4.51	1.4	170	3.9	320
Cr Kα	5.41	2.3	340	5.6	460
Mn Kα	5.90	1.7	260	8.4	530
Major elements: quantify to 1.0% ± 0.1% k > 10 indicates successful quantification					
Na Kα	1.04	140	57	170	110
Mg Kα	1.25	140	180	260	350
Al Kα	1.49	x	x	220	850
Si Kα	1.74	x	x	x	x
Ca Kα	3.69	240	11000	210	19000
Fe Kα	6.40	x	x	560	53000

x = when an element is present in significant quantity in the matrix, k can't be calculated by the method we used.

Case 1: Detection/quantification of astrobiologically significant elements on a rocky planet (e.g., Mars) or rocky planetismal (e.g., Phobos, Demos).

Case 2: Significance Level k for Biogenically important elements on an icy planetesimal (Europa), *30 mCi ^{244}Cm source .)*

Significance Level k of Peaks in Ice matrix with 30 mCi Cm244 source, run time 10000 seconds

Line	Energy (KeV)	Weight %	Significance Level *k*
C Ka	0.282	0.1%	8.3
N Ka	0.392	0.1%	18
Na Kα	1.04	0.1%	23
Mg Ka	1.25	0.1%	33
P Kα	2.02	0.1%	80
S Kα	2.31	0.1%	72
Cl Kα	2.62	0.1%	57

Case 2 Biogenic Elements on an icy planetesimal (e.g., Europa).

Case 3: Significance Level k of peaks on zero bkg. filter with 30 mCi ^{244}Cm source, 10^5 sec.
k > 2 = detection, k > 10 = quantification

Line	Energy (KeV)	concentration	Significance Level *k*
C Kα	0.282	1 microbe / 100X100 μm pixel	27.743
N Kα	0.392	1 microbe / 100X100 μm pixel	11.456

Case 3: Biogenically significant elements C, N on a zero background substrate on which Europa ice has been lyophilized.

Discussion:

Monte Carlo simulations of ^{244}Cm PIXE fluorescence of the biogenic elements in rock and water ice matrices demonstrate the value of this technique to landed science on Ocean Worlds. Development and NRC licensing of these sources should be made a priority for Planetary Science missions. ^{244}Cm can be procured through Eckert and Ziegler Isotope Products, but licensing and testing will require a commitment from the agency and the creation of dedicate facilities for source development and testing for fight.

References: [1]. Rieder, R., et al. (2003) *JGR-Planets, No. E12, 8066, doi:10.1029/2003JE002150, 2003.* [2]. Gellert, R., et al. (2006). *J. Geophys. Res. 111, E02S05, doi: 10/1029/2005JE002555 (2006).* [3]. Economou, T. (2011) http://www.intechopen.com/books/radioisotopes-applications-in-physical-sciences. [4]. Radchenko, V. et al. (2000). *Applied Radiation and Isotopes* 53 (2000), 821-824. [5] D.F. Blake et al. , IPM 2016 #2274. [6] T.Schoonjans et al. (2012) Spectrachim. Acta B, 70, 10-23. [7] Solé V.A. et al. (2007) *Spectrochim. Acta Part B, 62*, 63-68. [8] Agostinelli, S. et al. (2003), "GEANT4 – a simulation toolkit." *Nucl. Instr. and Methods in Phys. Research A*, **506**, 250-303.

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