## How to obtain a high-SNR EPMA map?: complementary roles of high-resolution imaging and subsequent binning

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Chemical imaging is a common technique to analyze heterogeneous materials. For example in petrology, X-ray mapping with an electron probe microanalyzer (EPMA) visualizes chemical heterogeneities (so-called texture) in rocks. X-ray mapping measures signals of X-rays, and higher signal-to-noise ratio (SNR) improves clarity of heterogeneities, which is generally achieved by increasing probe current and/or dwell time of pixels (e.g., Hiroi et al., 1997). An appropriate probe current is determined by sample damages, upper limits of the detectors, and dead time. Dwell time is also determined by sample damages and available analysis time. To acquire a high-SNR image at a reasonable time, reducing total number of pixels is one plausible solution; however, spatial resolution shall become low. To overcome this trade-off, this study proposes a complementary use of short dwell time mapping and subsequent binning. The short dwell time mapping generates high-spatial-resolution and low-SNR images for observing fine textures, and the binning treatment converts the raw maps to low-spatial-resolution and high-SNR images for detecting small differences in chemistry. This approach is applicable to any imaging techniques, including EPMA, which count signals under a given dwell time.

Chemical imaging records counts of signals from multiple points, typically as orthogonal pixels. If expected counting rates are fixed for pixels, observed counts follow Poisson distribution. In EPMA mapping, this assumption is valid because counting rates of X-rays depend mostly on (1) chemistry and surface conditions of materials which are constant; and (2) on probe current, accelerating voltage, and sensitivity of detectors which are controlled by analysts. According to the reproducibility of Poisson distribution, an expected counting rate of a certain signal from a large pixel is identical to an arithmetic-mean of counting rates of the signal from pixels dividing the large pixel. In other word, a sum of counts from n-by-n pixels in m seconds each is equivalent to measuring a large pixel composed of the n-by-n pixels in mn<sup>2</sup> seconds. A simulation is performed on a 300-by-300 matrix of Mandelbrot set. Expected counts per second is set to squares of Mandelbrot set timed by 20. Then, a 300-by-300 map with 1 second of dwell time, and a 150-by-150 map with 4 seconds of dwell time are generated by Poisson random numbers. The 300-by-300 map has higher spatial resolutions and lower-SNR than the 150-by-150 maps. Let's consider a binning treatment of the 300-by-300 map, i.e. combining signal intensity of each 2-by-2 pixels into one superpixel. The binned 150-by-150 map is apparently comparable to the native 150-by-150 map. This means that high-SNR (low-resolution) image can be generated subsequently from high-resolution (low-SNR) image at an arbitrary degree, if necessary. A recommended binning method is sum rather than arithmetic mean so that binned values can be modeled with Poisson distributions. For example, if a pixel value is generated by Poisson distribution, its standard deviation can be approximated as square root of the pixel value.

The proposed approach has advantages in statistical analysis dealing with mapping data. For example, cluster analysis on a map classifies pixels into a given number of groups based on similarity of pixel values. Performance of cluster analysis, such as precision and smoothness, may improve by obtaining high-SNR of pixels through binning which separates signal peaks representing different groups. Note that benefit of

sum as a binning method appears in cluster analysis because Poisson-distribution-based clustering algorithm (Witten, 2011) is applicable. If pixel values follow Poisson distributions, this algorism is more appropriate and provides better performance than k-means cluster algorism (MacQueen, 1967) which is a simple and common algorism used in variety of fields (Yasumoto et al. 2018).

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