

# The Importance of Sample Loading in Commercial Through-Belt PGNAA Applications.

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## Summary

The analytical physics of prompt gamma emission due to neutron interactions with matter has been well understood to good accuracy since the 1960's and applied to excellent effect in laboratory situations.

It is however naïve to underestimate the engineering challenges and subsequent complications of the analysis technique that arise when moving this well proven technique from the laboratory into a through-conveyor-belt industrial commercial mode of analysis.

This paper provides a word of caution to PGNAA users by demonstrating that an analysis based on simple linear combinations of elemental spectra is not sufficient to produce an accurate result over a wide range of compositions and sample loadings on the conveyor belt.

There are complex non-linear spectral effects introduced by a changing sample load and profile on the conveyor belt. This is primarily due to the changing thermalisation pattern of the neutrons, caused by the changing geometrical relationship between the source, sample, and detector as sample loading on the belt increases. Further, sample self-absorption also affects the thermalisation distribution within the material being analysed.

The conclusion is that detailed understanding and modelling of these non-linear effects is required to provide both unbiased and precise prediction of elemental composition of the sample compared to the presumption that linear regression techniques, such as library least squares, will suffice.

## Introduction

The physics of neutron interactions with matter, and in particular prompt gamma photon emissions from interactions of neutrons with atomic nuclei, have been understood quantitatively and to good accuracy since the 1960's. Comprehensive tables of such interactions are readily available from organisations like the International Atomic Energy Agency (<https://www-nds.iaea.org/pgaa/> )

The subsequent interaction of those photons with matter, including capture and measurement in a scintillation detector are also well characterised and understood ("Gamma-Ray Interactions with Matter" G. Nelson, D. Reilly.)

The translation of PGNAA technology from a laboratory situation to an industrial through-belt context sounds straightforward but is in fact complicated by several factors:

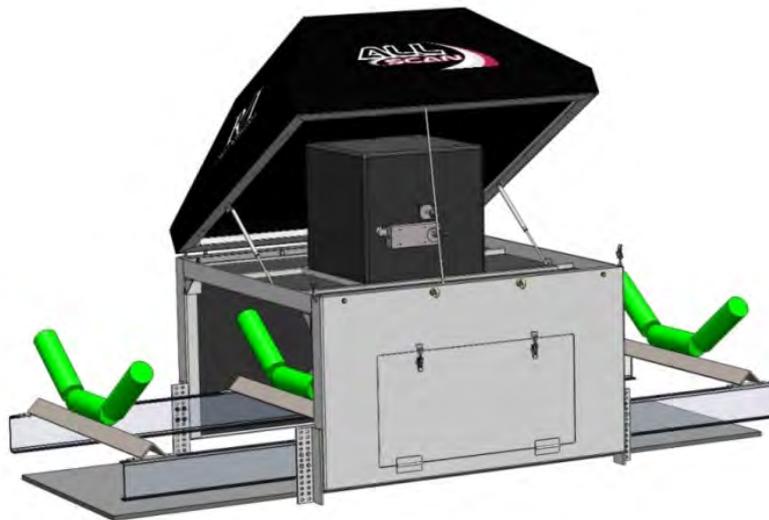
- a) The move from a carefully controlled environment to a dynamic changeable environment,
- b) The move from a carefully controlled geometry to a dynamic changing sample geometry and amount.

The traditional paradigm that combinations of fixed elemental "library" spectra can add up through linear combinations to the observed spectrum is often true under controlled calibrated laboratory conditions; however, there are many assumptions built into this statement that need to be questioned when you move to a through belt application.

This paper explores a few of the engineering realities of typical commercial through-belt configurations and exposes several complex factors that subsequently arise in the analysis of the prompt gamma spectrum to produce an estimate of the elemental composition of the sample.

## Comparison of commercial through belt design and laboratory setup.

Figure 1 shows a typical commercial through-belt PGNAA configuration which is superficially like a laboratory PGNAA setup. In both cases a sample exposed to a neutron flux produces prompt gamma photons that are counted by a photomultiplier tube. The gamma energy spectrum produced is analysed to determine sample composition, given pre-existing knowledge of how each atomic element produces a characteristic spread of prompt gamma photon energies.



**Figure 1: Typical through-belt PGNAA configuration (source at top, detector and belt not shown)**

The measurement process is straightforward – a high speed neutron flux from a source (typically Californium 232) is slowed sufficiently (thermalised) so that it will interact with atomic nuclei in the sample on the conveyor belt with a high probability of generating prompt gamma photons. Some of the photons are emitted towards the detector under the belt where they are counted.

Because each atomic element type in the sample has a characteristic gamma photon energy response to neutron activation of its nucleus, knowledge of the entire energy spectrum of the gamma photons can in principle be deconvoluted to estimate how much of each element is in the sample itself.

One of the issues arising in on-belt PGNAA applications is that these nuclear processes are essentially random within the analytical space. Neutrons will not only interact with the sample but with matter immediately around the sample (belt, idlers, support structure, detector and so on). So the spectrum is contaminated by photons that do not arise from the sample itself.

Also, many gamma photons emitted by the sample will not even be in a direction to interact with the detector, or they may interact with other matter (sample, sample holder, detector casing) before reaching the detector. High energy photons (above 1.1 MeV) may undergo pair production interactions.

In total, there are plenty of opportunities for complexity in the analysis to arise from what in principle appears to be a straightforward sequence of events.

In a laboratory situation, most of these complicating factors are strictly controlled by controlling the geometry and physical construction of the experimental setup. The gamma spectrum produced is by design tightly linked to a single tightly controlled sample volume constrained to a fixed known geometry.

By contrast in a commercial situation, specifically a through-belt PGNAA application, while the detector/source relationship is fixed, it is obvious that the geometry of the sample on the conveyor belt cannot be controlled to the same extent as in the laboratory environment.

Typically, the sample on the belt to be analysed is also particulate in nature, of varying size distribution, void packing, loading rate (kilograms per metre of belt) and profile (shape of sample on belt). In addition, the belt may drift slightly on the idlers as it runs and will change in appearance and shape depending on belt tension and sample loading. All these factors will alter the resulting PGNAA spectrum in subtle and not so subtle ways, even when the elemental composition on the belt remains unchanged.

Finally, there is a commercial context in which these PGNAA gauges are supplied. They must be affordable for the duty they perform to the client. It is simply not possible to justify the cost of large highly specialised detector crystals and photomultiplier tubes used in a typical laboratory standards setup. The processing of gamma pulses from such detectors must be done in real time in a commercial gauge – in contrast measurement in a laboratory situation can be recorded and processed later.

The reality is that lower cost scintillation detectors (typically Sodium Iodide or variants thereof) must be used in a commercial context but each brings with it their own challenges and limitations in spectral accuracy (pulse pile-up discrimination and rejection and so on.), compared to the more expensive germanium detector configurations.

### [The Effect of Sample Loading Geometry Relative to Source and Detector Position](#)

In a laboratory situation with relatively small sample sizes of well-defined geometry, the measured spectrum is by design going to move up and down in a linear scale in proportion to the sample mass of each element – that is a direct linear relationship between the mass of an element and its spectral response. The empirical setup is designed and instruments chosen of extra high quality (and cost!) to make this linear relationship a reality.

The engineering reality of a commercial PGNAA application means that some of these key laboratory design factors and equipment choices just cannot be met in practice. Of most concern is the fact that as sample load increases in a through-belt gauge, the geometric relationship between source, sample and detector of necessity, must also change as a function of sample loading. This does not happen in a typical laboratory setup where sample size is a strictly controlled variable.

Thus, this presumption of linearity of spectrum with sample mass may no longer actually hold. It is determining the correctness or not of this linearity presumption in through-belt PGNAA that is the main purpose of this paper.

## Measured Effect of Belt Loading of sample on Prompt Gamma Spectra

The distribution of prompt gamma count rate with energy, that is the spectrum from the multi-channel analyser (MCA) of course changes significantly with commodity. But it also changes significantly with changes in sample loading of that commodity on the belt.

Spectra collected from various loadings of different commodities will now be shown to illustrate the non-linear nature of some of the effects of belt loading geometry on spectral response.

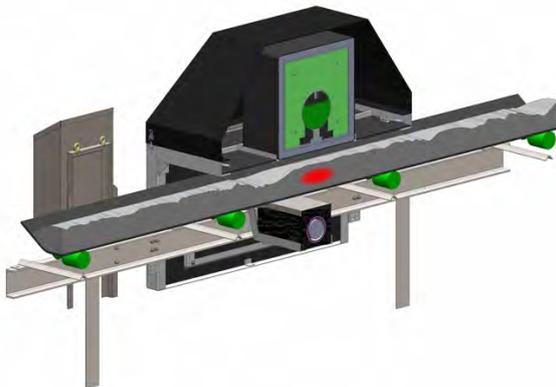
Almost pure iron ore ( $\text{Fe}_2\text{O}_3$ ) is used as a starting point in this demonstration because the spectrum is dominated by a single strong prompt gamma response element (iron) and is therefore relatively straightforward to analyse compared to complex mixtures of elements.

Figure 2 below shows a typical setup of sample bags loaded in a test rig.

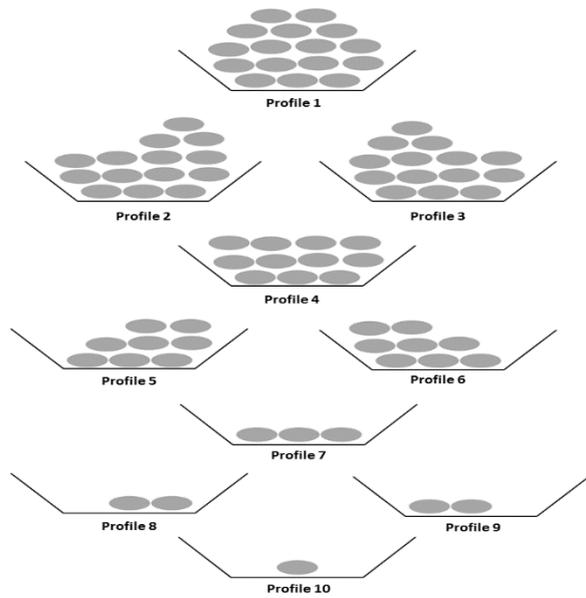


**Figure 2: Typical loading of samples in beam path of a through-belt PGNAA gauge.**

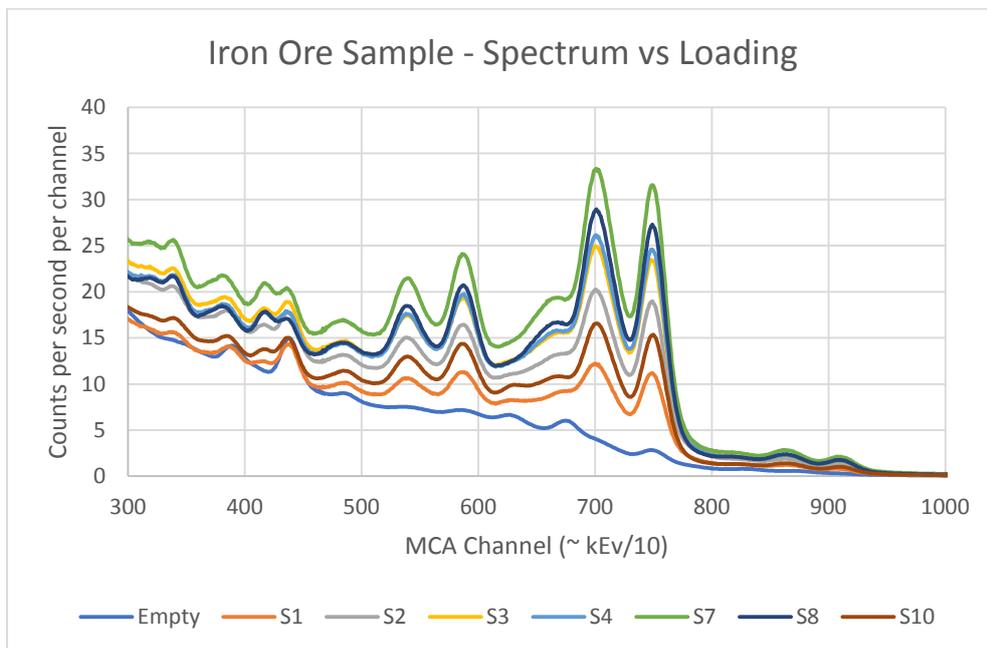
Identical bags (composition and weight) of an Iron Ore sample were progressively laid on top of each other on the conveyor belt in the test rig environment (see figure 2a,3), and the resulting spectra collected (as shown in Figure 4).



**Figure 2 a): Cross Section of Analyser indicating primary activation of sample material**



**Figure 3: Changing Sample Geometry due to Increased Sample Loading**



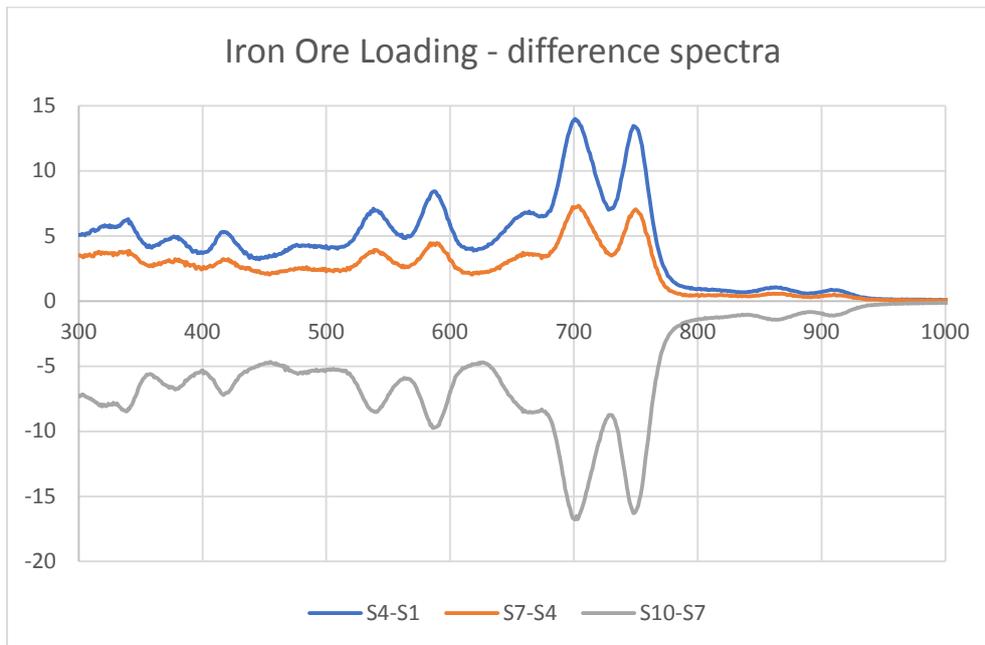
**Figure 4: Spectrum Response to Various Loadings of Iron Ore**

The first point to note is the existence of a significant empty belt spectrum that has nothing at all to do with the sample. This is the result of neutrons interacting with the immediate structure of the test rig itself – the conveyor belt, idlers, support structure, detector, and so on.

This means that the addition of a sample will result in a mixed spectrum of both sample and this surrounding environment, and will not just be a pure spectrum due to the sample itself.

Secondly, increasing from low numbers of bags we see the expected increasing sample response (increased count rates with increased sample mass). But this effect reaches a maximum response at 7 bags of sample (S7) and then starts to decline again. After a certain point adding more sample becomes self-defeating!

In Figure 5, the difference in spectra (S4-S1, S7-S4, and S10-S7) are plotted. As such, each differenced spectra is the spectral response caused by adding exactly 3 bags of identical sample, but not each set of three bags is added from a different initial starting condition.



**Figure 5: Spectral difference with iron ore loading – each difference represents the addition of 3 bags of identical material to the analyser.**

A naïve interpretation of spectral linearity would propose that these three difference spectra should all be identical. Clearly from figure 5, they are not. Adding three bags from a low loading (S4-S1) generates the largest count rate response. Adding another three bags (S7-S4) generates a similar but smaller response. The spectra do not measure the same scale of response even though three identical bags of sample have been added in both cases. Finally, the S10-S7 sample has generated a negative response. The count rate from the sample has gone down even though the amount of sample has increased.

These results indicate that just defining a simple model where a certain percent change in spectral count corresponds to a certain percent change in sample mass will in fact be biased and incorrect if applied over a wide range of sample loading on the belt.

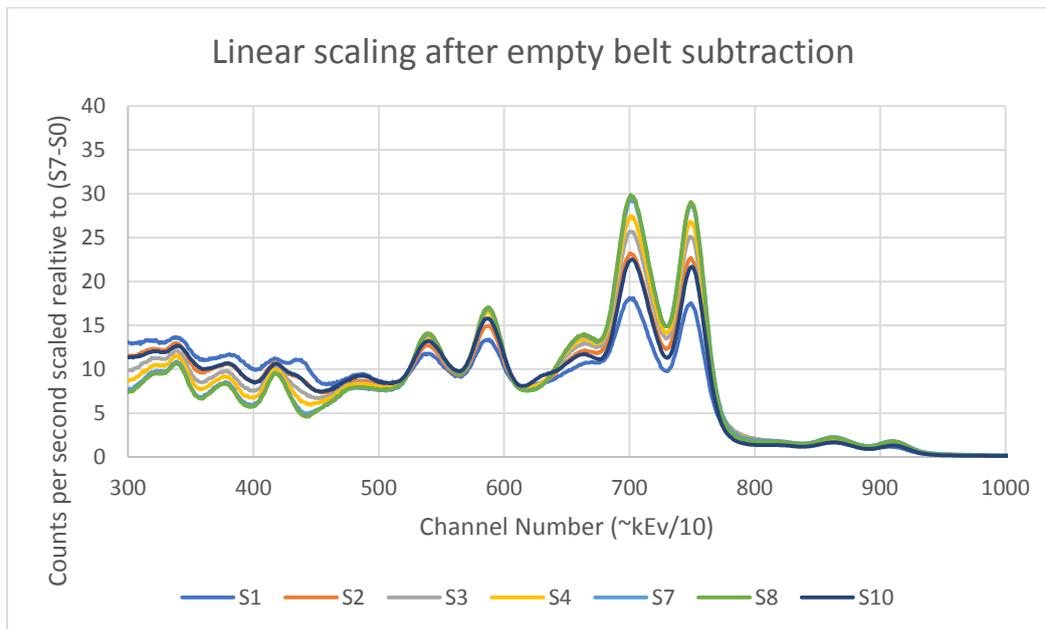
It is now demonstrated that things are even more complicated than this non-linear spectral scale factor with sample mass – not only can the same changes in identical sample result in different scales of spectral response, the issue of whether these responses are linearly related by channel energy is now explored.

Even if the scale of the spectral response with sample loading changes, it might still be argued that the spectra themselves should scale as a linear function with gamma energy, even if the scale factor itself is non-linear with respect to sample mass.

By this assumption, a single basis spectrum (SIO) should represent the sample, meaning all the spectra in Figure 4, should be simple scalar multiples of this basis spectra (less of course the non-sample contribution due to the empty belt environment). That is,  $S(n)-S(0) = a(n)*SIO$ , so the spectra are related by a linear factor  $a(n)$  even though  $a(n)$  itself will be a nonlinear function of  $n$  as we have already determined to be the case from Figure 5.

In this analysis the basis spectra (SIO) is taken as  $S7-S0$ , (i.e.  $a(7)=1$ ). If all the spectra in Figure 5 are related in a linear manner, it follows that any one of them may be chosen as the basis vector, and sample S7 is chosen because 7 bags of sample gave the highest count rate and most accurate response for the sample.

This linear fitting procedure is now applied to the spectra to see how accurately each spectrum in Figure 4 scales to this  $S7-S0$  base. The result is shown in Figure 6.

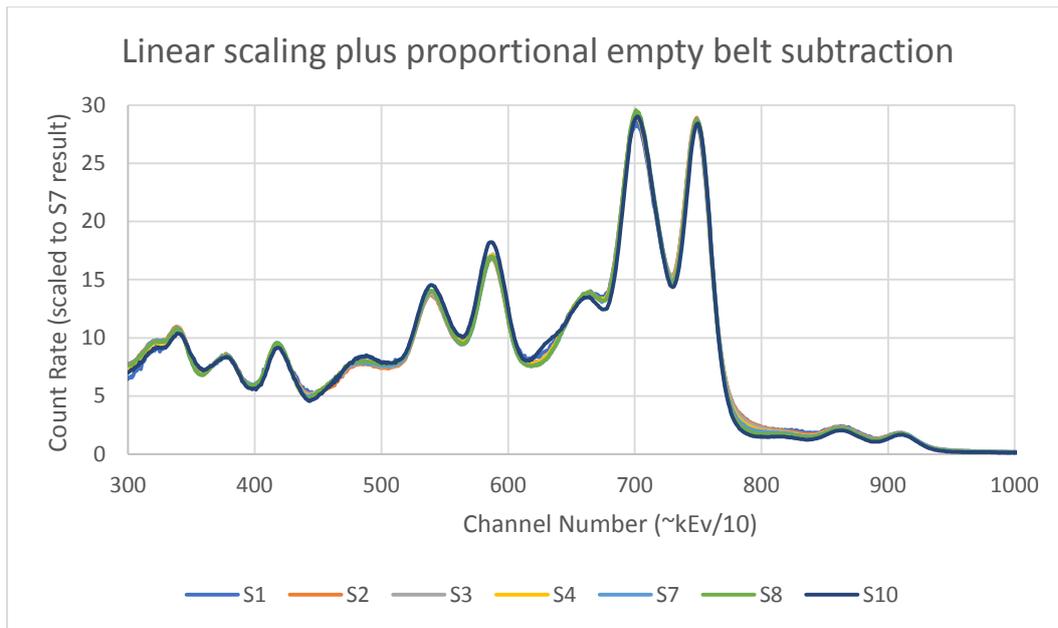


**Figure 6: Linear scaling of spectra after constant empty belt subtraction**

If the linearity hypothesis was correct, all these curves should be coincident with each other, and clearly, they are not.

Perhaps though, this linear analysis does not correctly account for the fact that as sample is added, because the neutrons being absorbed in the sample are no longer available to contribute to the empty belt spectrum; it is perhaps not correct to just subtract the empty belt spectrum,  $S0$  in total, but rather a linear proportion of the empty belt should be subtracted as sample loading increases.

Shown in Figure 7, this alternative linear model results in a slightly different response where  $S(n)-b(n)*S0 = a(n)*SIO$ . That is the spectrum  $S(n)$  of a sample at loading  $(n)$  is a linear combination of both the empty belt spectrum  $S(0)$  and the SIO sample basis spectrum.

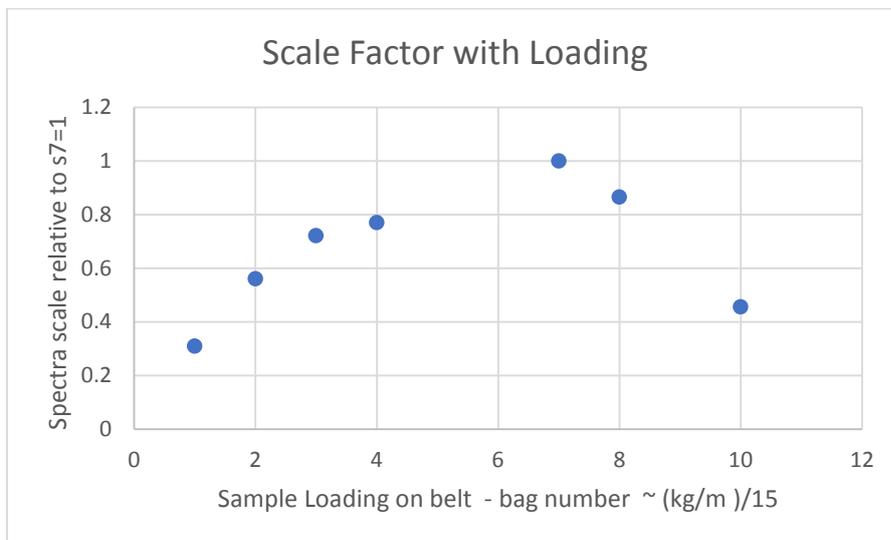


**Figure 7: Linear scaling after a proportionate subtraction of the empty belt spectrum**

While Figure 7 certainly shows a much-improved result to Figure 6, close inspection reveals small but significant differences remain between all the scaled spectra. If the average result in Figure 7 was used as a linear basis spectra for the sample (almost pure iron in this case), any linear fitting process would remain slightly biased with sample loading by these observed differences.

While these differences look tiny, it should also be remembered that this is just the spectral response to loading of essentially just one element (Fe) in an almost pure iron ore sample. Fe also happens to have a quite strong nuclear cross section, and other elements have very much smaller spectral responses to that in Figure 7. So much smaller in fact (as will be shown later) that even the tiny differences apparent in Figure 7 become significant in scale to these other elemental fits.

Finally, from the model used for figure 7, the scaling coefficients with loading,  $a(n)$ , can now be quantified in graphical form (Figure 8). This shows how a proportional change in sample loading results in a significantly non-linear response in the scale of the change in spectra.



**Figure 8: Scale response with loading showing an obvious non- linear response**

The conclusion is that linear modelling of spectra is only an approximate way to analyse spectral response vs gamma energy, and incorrect for working out the scale of correction needed under a wide range of sample loading circumstances.

### Why are Linear Spectral Models Approximate Solutions for Elemental Analysis of through-belt PGNAA spectra.

It is now worth spending some time examining exactly why linear elemental models can only approximate the spectral response of a commercial through-belt PGNAA gauge. The reasons roughly split into two causes:

- a) Internal instrumentation errors in the MCA pulse train analysis used to produce the spectrum.
- b) Geometry changes in the relationship between source, sample and detector as sample loading and commodity composition changes.

The main purpose of this paper is to bring attention to the realities of PGNAA analysis arising from the second factor. However, a brief aside is first made to internal MCA instrumentation errors to ensure it is realised that geometry changes with loading are not the only source of problems needing redress in a more “correct” non-linear analysis method.

The commercial MCA must cumulate spectral histograms with energy in real-time because clients need to know what is on their conveyor belts now, not in eight hours’ time. The sample rate from a photomultiplier tube is high (typically 100 MHz) so there is very little computational time available to detect and subtract a prompt gamma photon from background noise and ensure it is not contaminated by factors like pulse pile-up.

In practice, residual MCA instrument error often arises from three primary causes:

1. Inadequate estimate of the baseline signal which is subtracted from the area of the pulse signal to give the measured photon energy.
2. Non-linearity in the pulse amplification cascade/HV supply interaction in the dynode chain of the photomultiplier tube (PMT).
3. Incomplete detection of and rejection (or correction) where the electrical pulses from two gamma photons overlap each other (pulse pile-up)

While great efforts are of course made by MCA manufacturers to minimise these effects, it appears a fact of life that all commercial MCA’s in the typical price range suited to the through-belt PGNAA gauge market suffer from these residual errors to a smaller or greater degree.

The first two problems (poor baseline estimation and PMT/HV interactions) generally show up as a slight non-linearity between channel number and gamma energy. Provided these non-linearity’s are stable over time, a fixed energy calibration on the MCA can often be used to mitigate most of these problems.

Pulse pileup is where two separate gamma photons arrive almost simultaneously, but overlap in the detector circuitry. Rather than correctly detect and count 2 separate gammas, only one gamma is recording at the sum of the two energies. This misdetection remains a challenge for most MCA suppliers: Some MCA’s prove better than others, but none yet seem to have demonstrated effectively complete elimination of the problem.

The more of these “false” high energy events are counted as real, or not detected and rejected or corrected, the less accurate is the assumption that a complete sample spectrum can be reduced to a linear combination of elemental spectra.

## Spectral Non-linear effects due to Instrument Geometry

Following this brief aside, attention is now turned to the main point of the paper.

The increasing sample load on the belt of a PGNAA gauge presents a changing geometrical relationship between the source, the sample, and the detector, as shown below in Figure 8a.

The source is fixed above the belt and emits neutrons down towards the sample on the belt, which absorb the neutrons and generates prompt gamma photons, some of which keep travelling down to the detector mounted underneath the belt.

Even though the belt, source and detector remain in a fixed relationship, the pile of sample on the belt gets bigger as sample load (kilogram per metre of belt) increases. From pure geometry, on average the sample is closer to the source, and further from the detector as load increases.

This issue conflates with two additional factors to produce non-linear effects in elemental spectral combinations that essentially make simple linear elemental “library” spectra based approaches prone to bias and inaccurate fitting. This conflation is also illustrated in Figure 8a below.

The first issue is the pragmatic one of neutron thermalisation. The typical neutron source used in a commercial PGNAA through belt gauge (Californium 232) emits neutrons at too high a velocity to effectively interact with atomic nuclei to produce prompt gamma photons. The neutrons first must be slowed down or “thermalised” by the sample itself before prompt gamma interactions with nuclei can occur. In some cases, a pre-thermalisation assist can be given with a fixed material layer in the collimation path, but the issue remains: A changing sample amount changes the position of the thermalised neutron flux relative to the equipment, sample, and detector.

The second issue is the existence of a background spectrum or empty belt spectrum – that is the prompt gammas produced due to neutrons that by chance do not interact with the sample, but rather the immediate structure of the gauge itself (conveyor belt, support structures and so on).



**Figure 8a: Movement in Sample Activation due to Increased Sample Loading**

If it was just a matter of the neutrons that interact with the sample now being denied the opportunity to interact with the gauge support structure, then it would be a simple matter of a linear subtraction of some proportion of the known empty belt spectrum, to be left with a “pure” sample spectrum.

Unfortunately, this is typically not the case. As the sample mass increases, the thermalisation zone also rises in height with the rise in centre of mass of the sample. That is, the distance away from the source where most neutrons have slowed down enough to be a “thermal” neutron is reduced as sample mass increases. The rate at which this happens is dependent also on the commodity on the belt – some atomic elements are much more efficient at thermalising neutrons than others.

This now means different parts of the gauge superstructure (belt, idlers, supports and detector) receive different amounts of the remaining neutron flux not absorbed by the sample because the thermalisation zone itself has shifted.

So, it is simply not possible to assume the empty belt spectrum only changes in scale but that it must also change in spectral shape with gamma energy as more and more sample is added.

This is now a non-linear problem of determining what spectral changes are due to sample addition, and what are due to a changing environmental response to that sample addition. It is much more challenging to solve.

The third issue with the through-belt PGNAA spectrum is the fact that as sample mass rises, the prompt gammas produced in the top of the sample will be progressively further away from the detector, and will be subject to absorption by the sample layers below it before reaching the detector.

An otherwise equal sample sub-mass further up in the sample compared to further down near the belt will not only produce fewer prompt gammas per kilogram per neutron flux, but also a lower proportion of low energy gammas compared to high energy gammas due to self-absorption within the sample will arrive at the detector.

Finally, as the sample mass increases even further, the thermalisation zone rises preferentially even further into the top layer sample and self-absorption and distance to detector effects means adding sample perversely results in a reduced count-rate by the sample at the detector, whereas at lower loadings the rate was increasing with sample mass.

### Spectral Non-Linearity Accruing from Commodity Composition and Sample Loading

As a final illustration of these effects, we will look at the addition of a fixed amount of an element on top of base sample loadings of different commodities (coal, iron ore, cement, and empty belt)

In each case the spectral result from a base load of a fixed commodity composition is compared to the same load but with addition of a small known identical mass of a sample element.

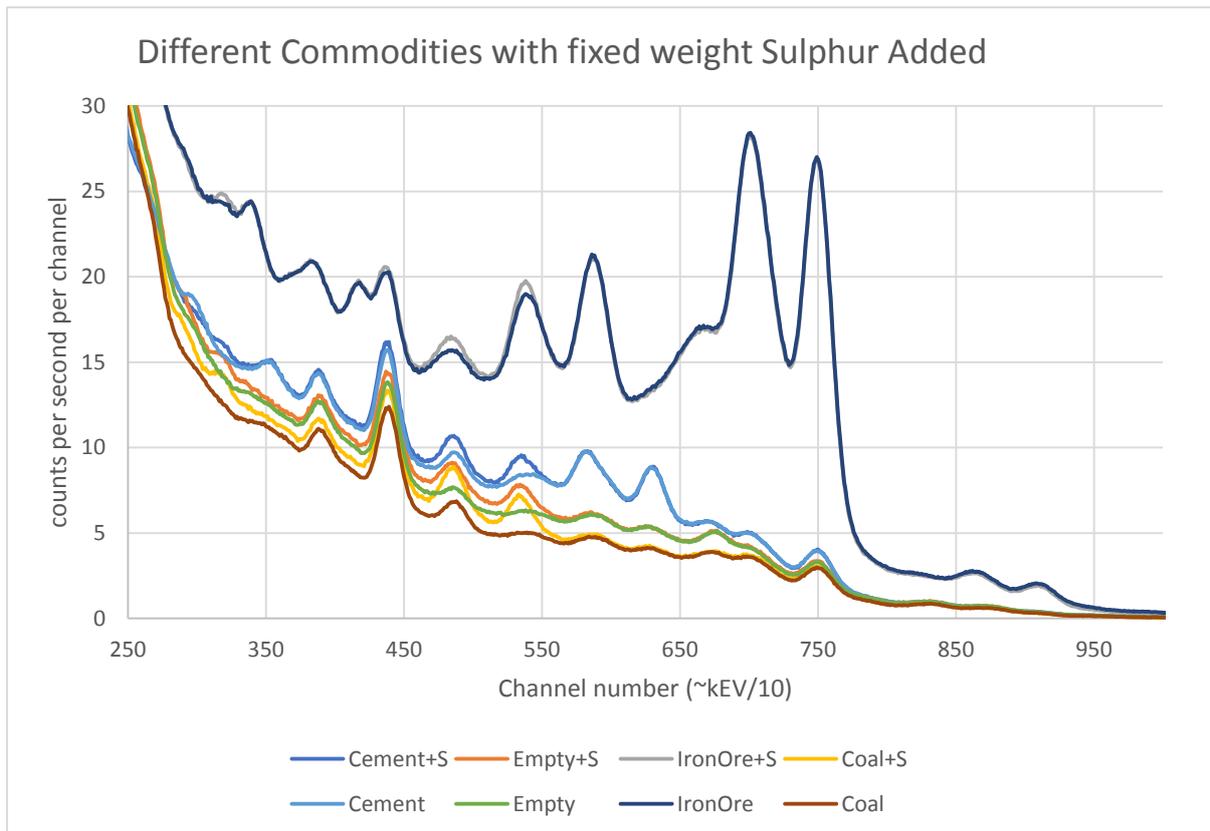
An average case is chosen. In this case 0.4 kg/m of Sulphur is added to each base load of 30kg/m Coal, 45 kg/m of typical cement plant fed, and 45 kg/m of Iron ore. Sulphur is chosen because it is an element of moderate or mid-range cross-section response to neutron activation compared to other common elements.

So, the approximate changes in elemental composition are about 1.3% w/w addition of sulphur for coal, and 0.9% w/w addition for cement and iron ore.

Finally, a comparison is made with the response of the same 0.4 kg/m of sulphur placed on an empty belt.

If the resulting data set of spectra could indeed be described by pure linear models of spectra, then an identical change in spectra should be observed in each case where the fixed amount of sulphur is added in the same geometrical relationship for each commodity.

Figure 9 below illustrates the actual results achieved.



**Figure 9: Different commodity base loads with fixed Sulphur addition.**

The first point to be made from Figure 9 is the wide range of spectral signatures observed due to the variation in elemental composition of the different “typical” commodity base compositions.

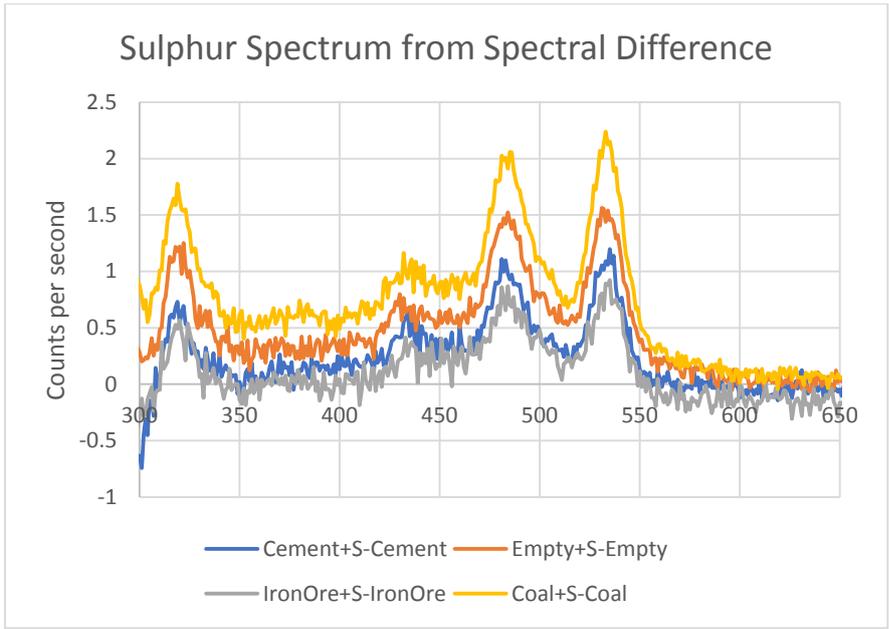
The second point is that (as per previous results) there already exists a significant spectrum due the empty belt before any sample at all is added.

The final point is that even with an approximately 1% w/w additive of an element with a moderately strong nuclear cross section for neutron absorption, the added Sulphur only makes very small change to the spectrum (of the order of a count per second per channel), for a given source strength.

This small change would indicate that even small deviations in linearity of response with commodity and loading are going to have significant impact in biasing the accuracy of elemental composition estimation at the one percent w/w level.

Sulphur was chosen in part because its elemental spectral peaks have by PGNAA standards a quite good separation from the positions of other elemental peaks so small spectral changes can be seen by eye, and in part because it has “typical” mid-range cross-section to judge response level.

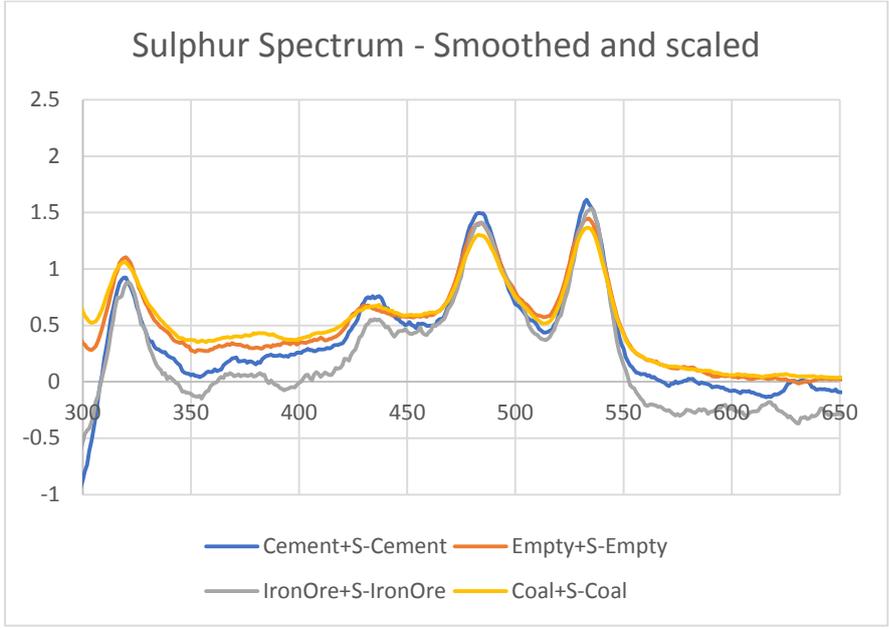
To expose the effect of just a small Sulphur addition on the change in spectrum, Figure 10 presents the difference in spectrum between the commodity plus sulphur compared to the commodity itself.



**Figure 10 – Presuming linearity, spectral difference to reveal just the effect of sulphur**

The first point is that even though exactly the same amount of sulphur was added in each case, the responses are quite different in scale as explained above due to each commodity and sample load change also changing the pattern of thermalisation of the neutrons. This is exacerbated by the different self-absorption effects accruing to a given depth of each different commodity.

This non-linear scale factor response can be further quantitatively explored by trying to scale each of these differenced sulphur spectra to fit the best spectral estimate for just sulphur alone (orange curve in figure 10). The curves below are also smoothed slightly to highlight differences between the estimated sulphur only spectrum from different commodity tests (assuming linear models)



**Figure 11: Scaled and smoothed estimates of sulphur spectrum assuming linear models**

The first point to note is that even with just a small amount of sulphur addition, the actual sulphur spectrum arrived at by differencing, does not in fact result in a single common sulphur (basis) spectra with gamma energy (channel number).

The second point is that extra non-linear scale factors are required for the sulphur spectrum to account for changes in neutron thermalisation and self-absorption due just to changes in commodity type and loading. For cement the best scale factor is 1.56, for iron ore it is 1.99, and for Coal it is 0.67. If there were no non-linear effects, all the spectra above would be identical and equal to the empty belt sulphur result, and the scale factors would all be one (indicating the non-changing nuclear cross section of Sulphur)

If the average sulphur spectral data from Figure 10 was used assuming just a linear model for sulphur, a cement commodity would underestimate the amount of change due to sulphur by 50%, by a factor of two in iron ore, and an overestimate by  $1/3^{\text{rd}}$  in coal.

Further, the fact that no one single spectra can be scaled to match the same sulphur addition for different commodities without leaving residuals in the peak fitting process, means that when fitting multiple elements by linear methods, the answers for other elements will also be further severely biased to compensate for this lack of goodness of fit.

In other words, if the scale factor needed to convert the Sulphur spectral change to sulphur composition change was purely just a function of its nuclear cross-section and not influenced by non-linear changes in neutron thermalisation, self-absorption and loading factors, all the spectra in figure 11 would be identical.

Since this is not the case, it proves the point that using linear spectral models leads to biased estimates of composition in practical through-belt PGNAAs applications. The same general results and trends are found to be true for all the different elements as it is for these sulphur results at different commodity types and belt loadings.

## Conclusion

The engineering realities of commercial through-belt PGNAAs gauges means that the assumption that elemental analysis in such situations can be achieved by linear combination models of fixed elemental reference spectra should be called into doubt.

The unavoidable non-linearity introduced by the changing geometrical relationship between source, sample and detector mean that while these linear presumptions may well work in a controllable laboratory PGNAAs context, they are in fact not well suited to commercial through-belt gauges

A correct solution requires a careful analysis and understanding of how the changing geometry with sample load and changing commodity base composition affects the position of the thermalization field of the neutron flux, coupled with the changing self-absorption affects due to the sample relative to the non-sample spectrum.

These corrections require key non-linear model adjustments for loading and self-absorption in conjunction with the known linear models to produce a precise and unbiased estimate of elemental composition under a wide range of loading and composition variability.

Without these corrections, purely linear models will at best be subject to bias and loss of precision in predicting elemental composition, and at worst they will become unreliable except perhaps when carefully linearized over a very narrow range of operating conditions of belt loading and commodity base composition.