Study on whether using an autosampler to sample metallurgical coke will reduce variation in its analysis

by Jamie Mathew Waters

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Study on whether automation of the sampling method for metallurgical coke will reduce variation in its quality analysis

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Declaration

This thesis is the result of the author's original research. It has been composed by the author and has not been previously submitted for examination which has led to the award of a degree.

Signed:

Date:

Acknowledgments

I would like to thank my project mentor, Dr Gregory Owen, for his guidance and advice while completing this project, I would also like to thank Andrew Thorn and all the other Lecturers at UWTSD Swansea that have taught me over the last four years and helped me through this period.

Thank you to my managers at Tatasteel who encouraged and supported the decision take on the BEng Material Science degree.

Lastly a very big thank you to my wife and two children for their patience, support and inspiration.

Abstract

Due to growing concerns over metallurgical coke quality, an autosampler was purchased by Tata steel, it was believed that a poor sampling regime was contributing to high variation in a coke quality measure called 'coke strength after reaction' (CSR). The lower specification of the CSR had been raised because of the concerns over quality, to facilitate this raise in specification, higher quality coals are needed in the coal blend used to make coke, costing the business approximately £8 million per annum. It was hoped that an autosampler along with another few initiatives would reduce variation and allow the specification to be raised back to its previous level, saving the business millions.

To try to evaluate the impact of the introduction of an autosampler, three months of coke quality data before installation of the autosampler were compared to two months (due to time constraints) after installation of the autosampler. The results of the data found that after two months of using the autosampler there was almost no change in CSR variation or mean. Implying that the sampling method was not a contributor to CSR variation. Six other quality measures (ash, moisture, mean size, cold strength-M40, Abrasion index-M10 and reactivity CRI) were also compared in the hope that a drop in standard deviation across a range of quality measures would demonstrate the influence of the autosampler.

Half of the six measures saw a drop in standard variation while the others increased. There are many variables in the coke making process and many of them will affect coke quality. Some of the measures saw a change in the mean suggesting that there had been changes in the process.

To enable proper measurement of the variation in the process the autosampler needs to be fully automated and the sample frequency needs to be increased from the four hourly increments in place currently.

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Introduction

In 2016 Tata steel announced that its UK assets were to be put up for sale, citing the reasons as global oversupply of steel, high cost of raw materials and electricity, the influx of cheap steel from countries such as china into Europe, a volatile currency and a weakness in the domestic market demand for steel (James, 2016).



Figure 1 - The Port Talbot steel works (Image: Matthew Horwood)

According to the BBC (2016) during this time Port Talbot steelworks was losing one million pounds per day. Tata had already announced 1050 job losses across the UK in January 2016 with 750 going at Port Talbot.

A rescue plan was eventually agreed, after the price of raw materials fell, steel prices increased and the value of the pound dropped after the UK voted to leave the European union. A rescue plan was drawn up which involved closing the £15 billion British steel pension and in exchange, Tata promised to keep Port Talbot's blast furnaces running until 2021, avoid any further redundancies and invest £1 billion into the plant over the next 10 years.

The job losses at Port Talbot affected all departments and many departments required a restructuring in order to cope with these job losses. For the laboratories, within the technical department, this meant a 20% reduction in its manning resulting in a decision to move from a 24-hour shift operation to a 12 hour day shift operation to cope with the reduction in manning. This reduction in resource impacted on the service the laboratories was able to provide.

The raw materials department within the laboratories is responsible for the collection, preparation and physical testing of all materials used in the heavy end of the steel making process including coal, metallurgical coke, sinter and all the materials used to make sinter and iron ore. Materials such as coal, coke and iron ore were sampled for analysis on a 24-hour basis. After the restructure this was no longer possible. New sampling regimes were agreed upon with the laboratory's customers namely the coke ovens, sinter plant and blast furnaces.

This project is going to focus on the sampling, preparation and testing of metallurgical coke and how sampling practices affect the physical and chemical analysis of the coke.

Historically, coke had been sampled at the coke ovens every four hours from a conveyor belt by a technician using a shovel to be taken back to the lab for analysis. Some tests were carried out on these individual samples and some tests were carried out on a 24-hour composite of these 4 hourly samples. The sampling technicians carrying out this sampling were fully trained in sampling techniques and understood the importance of representative sampling. Representative sampling means taking a small increment or sample from a larger bulk of material for measurement or analysis in an unbiased manner, any part of the lot or larger bulk must have an equal chance of being taken (ISO, 2001).

In the restructure the laboratories sampling technicians were placed on a two team, 12-hour day shift pattern, where one team worked four 12 hour shifts then had four days off and vice versa. It was agreed that the coke ovens arrange the sampling of the coke and the laboratories sampling technicians would collect the samples twice during their shift at 07:00 am and 17:00 pm. The sampling of the coke was assigned to the coke wharfman, the wharfman works in the vicinity of the sample point and his role is to oversee the coke as it is pushed from the ovens and loaded onto the conveyor belt system. The wharfman works for a contractor. Although trained how to take the sample correctly, the wharfman would not have the same sampling experience as the laboratory sampling technicians and would not understand the importance of representative sampling or the impact of poor sampling upon analysis.

In May 2017 the blast furnace team grew concerned, after a poor run of results of coke quality reported by the laboratories. The team were focusing on the coke strength after reactivity (CSR) and coke reactivity index (CRI) results in particular as these are important parameters used for coke quality. CSR and CRI results are obtained from one test. The blast furnace technical team were concerned about low results for CSR and the variation in the results from day to day. If the coke ovens had not made any changes to process parameters or coal blends, the results should not change much from one day to the next but large changes in results were happening regularly and falling below specification limits. An investigation into the CSR results swings was carried out by a member of the blast furnace technical team. The investigation

found that poor sampling practices could be a contributary factor. The amount of coke available for collection was often not as much as expected for 3 samples, there were occasions when there were no samples available at all, meaning the composite for analysis was made up of less samples and the conveyor belt was not stopped as regularly as it should have been.

Blast furnace 5 was due some extensive maintenance work in November 2018 designed to extend its life until 2025 and Port Talbot would rely on blast furnace 4 alone for iron making while this work was being carried out. The blast furnace technical team did not want to take any risks with coke quality during this time, as it is fundamental to blast furnace operations. An alternative coke in the form of Mitsibushi coke from Japan is used in the furnace around 22% and the percentage of its use could be increased. This would be an expensive option as approximately £23000 per week is spent on imported coke at present.

As poor sampling practices was considered a source of variation for inconsistent coke quality results, an autosampler was considered. An autosampler will be placed on the coke conveyor belt and mechanically take a sample from the belt effectively automating the process and remove any human error or bias.

Objectives

The objective of this project is to demonstrate that the use of an autosampler in coke sampling will reduce variation in coke analysis. Due to recent blast furnace instability, the aims of some coke quality measures have been changed to try improve the quality of the coke produced. To achieve these aims better quality coal is required at the coke ovens to produce the required quality of coke. Which comes at a cost. The level of variation seen in analysis has been deemed too high and it is theorised that the method of sampling is contributing to some of this variation.

Three months of data from a number of coke quality tests on coke sampled before installation of an autosampler is going to be compared to three months of data after installation of an autosampler. If it can be demonstrated that variation in the analysis is reduced sufficiently and is under control, then confidence in coke quality may be restored and the quality measure aims could be reduced leading to an opportunity for significant cost savings for the business.

Timing Plan

To keep track of the project objectives and to make sure they were met on time a Gannt chart was used see figure 2.

Most of the project objectives were met and the project mostly ran as planned. The biggest issues faced were delays caused by difficulties encountered when installing the autosampler. The delivery of the autosampler was set back a week because of a customs issue. Then during installation, the electrical engineers encountered difficulties in setting up the autosampler. For the autosampler to be run in automatic mode, sensors need to pick up whether there is actually material on the conveyor, these proved difficult to set up and were still not operational at the time of writing this report. The speed of the hammer on the autosampler also needed to be set correctly.

Then when the autosampler was ready a fault with the coke belt arose which took another week to resolve. It was hoped that data collection with the autosampler in use could start in week 49 – 2018 but this was delayed until week 6 – 2019. Originally three months' worth of autosampler data collection was planned but this had to be reduced to two months to ensure there was enough time left to analyse the data and write up the project.

Timeline Planning	Start:	6th July 2018						End:				December 17th 2018																		
What	Who																													
Short Term Tasks - Weeks		27	28	29	30	31	32	33	34	35	36	37	38	39	40	41	42	43	44	45	46	47	48	49	50	51	52	Status
Confirm that autosampler is to be purchased	JW																											Green
Write up project proposal	JW																											Green
Present at coke quality ISTR	JW																											Green
Hold meeting with stakeholders	JW																											Green
Get access to analysis database (ODBC)	JW																											Green
Collate data for manual sampling	JW																											Green
Meet with coke ovens technical to discuss autosampler	JW																											Green
Carry out preliminery research	JW																											Green
Hold autosampler planning meeting	JW																											Green
Analyse data collected	JW																											Green
Test autosampler	JW																											Green
Analyse some autosampler sames vs manual samples	JW																											Green
Present at senior management coke quality review	JW																											Green
Start sampling from autosampler (delayed)	JW																											Red
Write up interim viva	JW																											Green
Present interim viva	JW																											Green

Timeline Planning	Start:	1st January 2019						End:				13rd May 2019																		
What	Who																													
Short Term Tasks - Weeks		1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20							Status
Write up safe working procedure for autosampler	JW																											Green
Start writing up litriture review	JW																											Green
Analyse Manual sampling data	JW																											Green
Start using autosampler	JW																											Green
Analyse data collection from autosampler	JW																											Green
Get draft of project written up	JW																											Green
Finalise project report	JW																											Green
Write up Vivia and poster	JW																											Green

Figure 2 - Project Gannt Chart

Background

The Blast Furnace

Steel at Port Talbot is produced at the Basic Oxygen Steelmaking plant (BOS), the raw material used in this process is iron. Lundgren (2010) asserts, the most widely used method for producing iron is through a blast furnace. A blast furnace is charged through the top with iron bearing materials such as iron ore and sinter, metallurgical coke and fluxes or slag formers. The iron bearing materials are charged along with the fluxes in alternating layers with the coke. Preheated air (blast) and coal is injected near the bottom through tuyuers (water cooled copper pipes), the hot blast ignites and consumes the coal and coke producing a very hot flame (up to 2300°C). The hot gases produced rise up the furnace melting and reducing the iron ore. As oxygen in the burden is removed it reacts with the coke and produces carbon monoxide (CO) further fuelling the reduction of the iron ore in the burden. The voids caused by the burned coal and coke allows the burden in the furnace to descend. The fluxes react with impurities and produce a slag which along with the liquid iron seeps through the porous coke and is eventually tapped from the bottom, as the slag is less dense than the iron they do not mix (Lundgren, 2010).

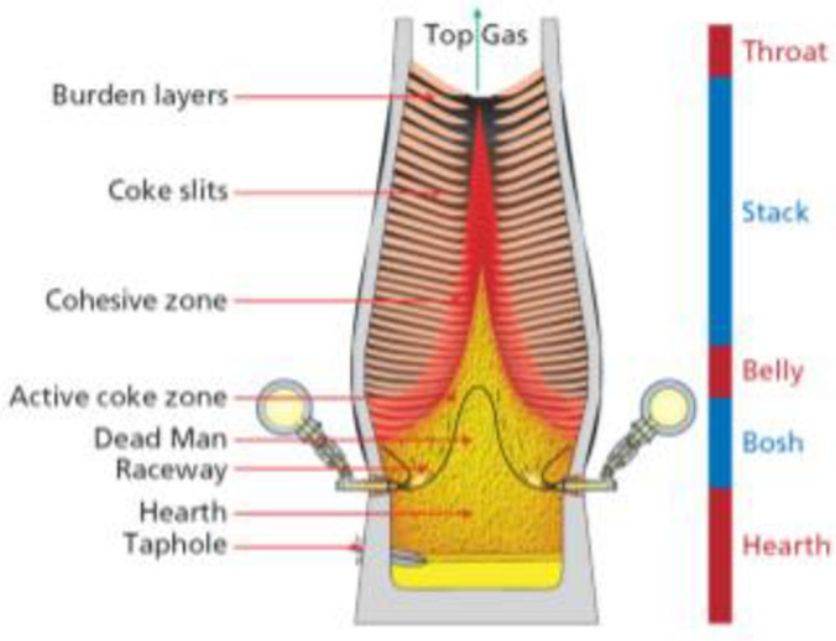


Figure 3 - The blast furnace (Geerdes, et al., 2009)

Haapakangas (2016) states that in the throat of the furnace any moisture present in the charged materials is driven off by the rising hot gas, as temperatures rise to 500 - 600°C the iron ore in the form of Hematite (Fe_2O_3) is reduced to Magnetite (Fe_3O_4). Descending a bit further as temperatures now rise to 700 - 900°C Magnetite reduces to Wustite (FeO) as the burden now reaches the cohesive zone the Wustite has reduced to metallic iron which starts to soften and melt at 1100 - 1400°C. Below the cohesive zone coke is the only solid raw material as temperatures raise over 1500°C.The active zone is a semi fluidised region in which liquids drip through voids between lumps of coke. The deadman is a stable region of coke through which liquid drips and the final stage of reduction takes place. By the time the burden reaches the bosh it is fully melted and reduced. The molten iron and slag collect at the hearth where it is cast via the tap hole.

Metallurgical coke



Figure 4 – Metallurgical Coke (Waters, 2018)

As Geerdes (2009) explains metallurgical coke is a carbonaceous material made up of 80% carbon, produced by the carbonisation of coal. This carbonisation is achieved by heating the coal up to 1100°C in the absence of oxygen. Coke is a solid material that stays solid at temperatures over 2000°C, it is porous, has a low ash content ~10% and high cold and hot strength. The importance of coke in blast furnace cannot be understated, as of all the materials fed into the furnace, it can have the biggest effect on blast furnace operations and hot metal quality. It is metallurgical cokes' excellent physical and chemical properties that allow it to carry out its roles within the furnace.

Li (2014) describes the roles of coke in a blast furnace.

- The oxygen in the hot blast reacts with the coke and produces carbon monoxide which then in turn reduces the iron ore.
- The coke acts as a fuel in the furnace providing the heat needed to melt the iron ore.
- Coke provides carbon for the carbonisation of the hot metal.
- Its porosity and permeability allow molten iron and slag to flow down the furnace and any gases to evenly rise to the top.
- As the only material that remains solid throughout the furnace, the coke acts a structure holding the full weight of the burden.

Due to the high cost of metallurgical coke there has been significant efforts made to reduce the amount of coke needed in the furnace. This has resulted in some replacement of coke with coal. The coal is used as a fuel and is injected into the furnace via the tuyeres. Approximately 300 Kg of coke and 200 Kg of coal are used per 1 tonne of hot metal produced. This reduction in the amount of coke used in the furnace means that the remaining coke is subjected to greater mechanical and chemical stresses. However, there are no other known materials that could replace coke as a support for the blast furnace burden (Díez, et al., 2002).

As a consequence, the quality of the coke used in the furnace becomes very important and it needs to have a number of properties to continue to allow gas permeation while simultaneously holding the burden up coke in the chemical and thermal environment of the furnace. Metallurgical coke needs to be resistant to degradation, abrasion and chemical attack, be of a certain size, have a specific chemical composition and mechanical strength.

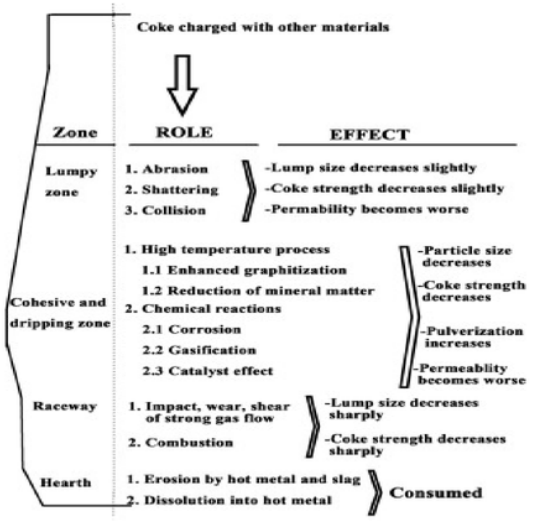


Figure 5 - The degradation process of coke in a blast furnace (Li, et al., 2014)

Therefore, there needs to be some measurements of these coke requirements to assess the quality of the coke being produced at Morfa coke ovens to enable the blast furnace to make predictions on its performance and to have the confidence in using it. There are a number of tests carried out at the laboratories in Port Talbot that help assess its quality.

Size Distribution

A large mean size is desirable with a low narrow size distribution, the coke is screened to control its size, most plants would consider 50 – 55 mm the optimum mean size. Fines in particular are detrimental as they will build up in the voids between the coke and reduce permeability. Fines can be described as the fraction of coke or any other material under 5mm. Fines are screened out of the coke and reused as fuel in the sinter plant. Fines can be caused by the breakdown and abrasion of the coke (Sharma, et al., 2013).

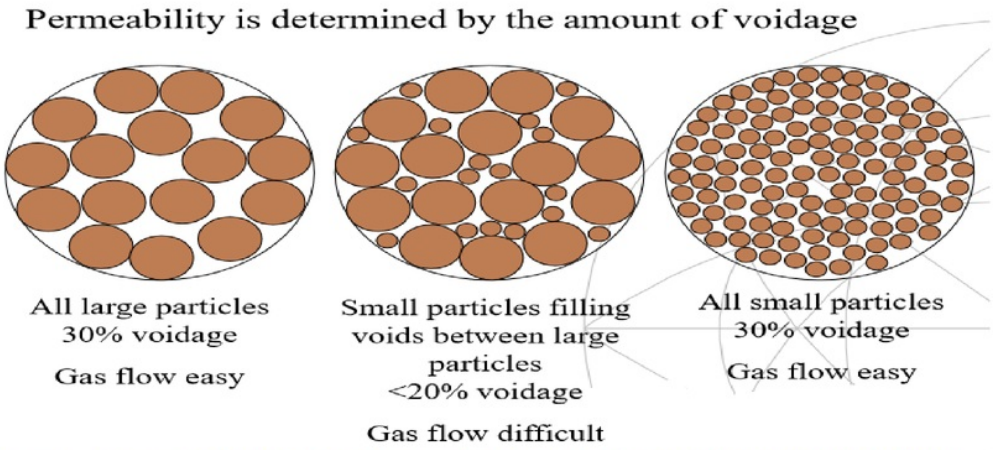


Figure 6 - Diagram explaining the importance of particle size in gas flow (Danieli Corus BV, 2009)

The laboratories measure the coke size before it is screened on plant, the size distribution is measured by passing approx. 50 Kg of coke over metal screens with round evenly distributed holes of specific size.

- +120 mm
- +100 mm
- +80 mm
- +60 mm
- +40 mm
- +25 mm
- +20 mm
- +10 mm
- -10 mm

The coke is collected in bins underneath the screens and the weight of each bin is measured. The particle size distribution can then be calculated.



Figure 7 – Coke particle sizing screens (Waters, 2018)

Mechanical Strength

After being produced at the coke ovens, the coke needs to be transported to the blast furnaces. During this transportation the coke is subjected to some mechanical stresses. It is dropped from conveyor belts onto stockpiles, it falls into bunkers before finally being charged into the furnace. This handling will cause some degradation of the coke due to abrasion and shock. This coke degradation process is called stabilisation, by the time the coke is charged into the furnace it is almost fully stabilised and the resulting coke is less likely to degrade any further and most of fines produced during stabilisation will be screened out (Sharma, et al., 2013).

The resistance to size degradation at room temperature is measured at Port Talbot by using the half micum test. The standard for which was developed in 1992 and is covered in the British Standard (BS) 1016. After the coke has been screened for size distribution measurement, a 25 Kg test portion is weighed out containing representations of each size fraction above 60mm from the gross sample. The sample is placed into a cylindrical drum, which is then rotated 100 times at 25 rpm. Paddles inside the drum lift the coke, as the paddle becomes vertical the coke drops to the bottom of the drum. This is meant to replicate the mechanical handling of the coke during transportation and charging. The coke is then screened again, the percentage of coke left over 40mm is reported as the M_{40} index and is the measure of strength in relation to degradation by volume breakage, the percentage of coke left under 10mm is reported as the M_{10} which is an abrasion index (Geerdes, et al., 2009).

Taken from (British Standards Institute, 1992)

The M_{40} and M_{10} are calculated as follows:

$$M_{40} = \frac{M4}{M3} \text{ x } 100$$

Where:

m_3= Mass of the test portion

m_4= Is the cumulative mass of coke (after rotation in the drum) retained on all sieves of aperture size equal to or greater than 40mm

$$M_{10} = \frac{(M1+M2)}{M3} \text{x} 100$$

Where:

m_1= is the mass of coke (after rotation in the drum) passing the sieve of aperture size10mm

m_2= is the mass of material lost during determination

m_3= is the mass of the test portion

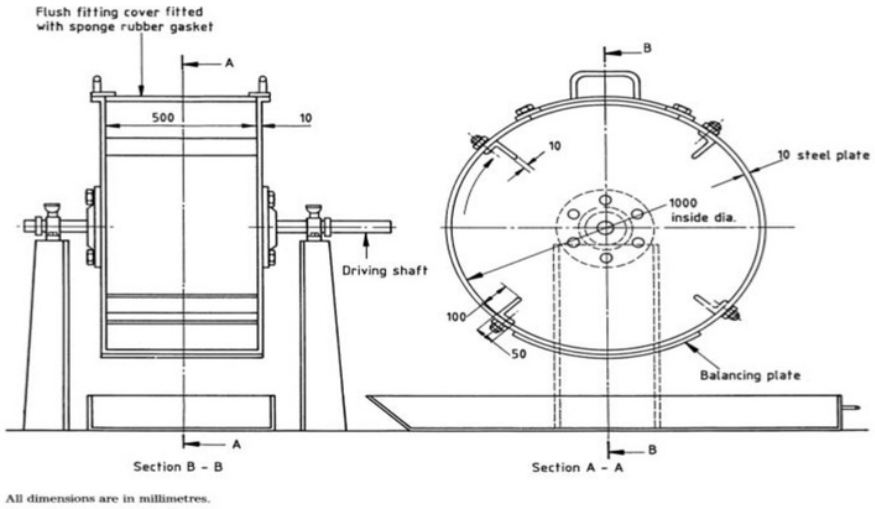


Figure 8 – Diagram of a Micum drum (British Standards Institute, 1992)



Figure 9 – The Micum Drum at the laboratories (Waters, 2018)

Proximate analysis

Moisture

Coke is cooled by quenching with water, after being pushed from a coke oven. A high moisture level in coke is undesirable as it will affect the coke weight and therefore the amount charged into the furnace, it also puts an added thermal load on the furnace. Once in the furnace the moisture will evaporate leaving less coke than desired. Fluctuations in moisture content in coke can result in erratic furnace conditions. To prevent this, measuring the moisture content of the coke is necessary (Han, 1993).

Coke moisture is tested according to BS 1016-102:2000 ISO 579:1999 standard and involves first crushing the coke sample in a jaw mill, then weighing out 1kg into a steel tray measuring 10cm x 10cm, the coke is spread across the tray and then put into a drying oven at 105°C for 3 hours. The weight lost is measured, the coke is then put back in the oven for another half hour and weighed again (British Standars Institute, 2000). This step is repeated until a constant mass is achieved. The moisture content can then be calculated as :

$$mT = \frac{(m1 - m2)}{(m3 - m2)}$$

Where: *m*T= is the total moisture expressed as a percentage

m1= is the mass of the tray plus the mass of the sample

$m2$= is the mass of the tray

$m3$= is the mass of the tray plus sample after heating

(British Standards Institute, 2000)

Ash

Coke ash is an important measurement as a high ash content increases blast furnace fuel rates and decreases hot metal productivity. The higher the ash content the less carbon content in the coke, it also lowers iron temperature and means more fluxes are required to melt the ash which increases slag volume and energy usage (Lundgren, 2010). Ash is gravimetrically measured by weighing 1 gram of pulverised, dry coke into a ceramic dish. The ceramic dish is placed into a furnace at 800°C for an hour where the carbon is burnt off leaving metallic oxide ash. The dish is cooled and weighed back. The weight left afterwards is the ash measurement.

Sulphur

Sulphur is detrimental to ironmaking, a high sulphur content in the iron means it has to be removed at the steelmaking stage. High sulphur levels also interrupt furnace operations and increase slag volume. When sulphur levels are high more limestone needs to be added, more coke then needs to be added at the expense of iron ore resulting in reduced iron production. Coke is the main source of sulphur in the blast furnace, sulphur levels in coke are dependent on the coals used to make the coke (Han, 1993).

Coke strength after reaction and Coke reactivity index (CSR/CRI)

Developed by the Nippon steel corporation in the 1970's as a way to evaluate coke hot strength. This test is an internationally accepted measure of coke quality and is designed to simulate the behaviour of coke under blast furnace conditions. The CRI index measures the percentage weight loss when exposed to 1,100°C for 2 hours, to provide an indication of coke resistance to temperatures typically found in the lower 2/3 of a blast furnace and CSR measures the potential of the coke to break into smaller sizes under a high temperature CO_2 environment that exists in lower 2/3 of the blast furnace (Díez, et al., 2002).

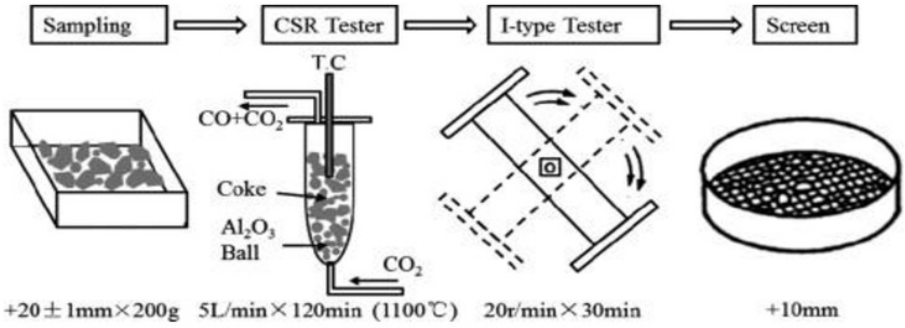


Figure 10 - Diagram of the CSR/CRI test (Li, et al., 2014)



Figure 11 - Pictures of the CSR/CRI testing equipment at Port Talbot (Waters, 2018)

The CRI is expressed as a percentage by mass and is given by the formula

$$CRI = 100 \times \frac{m_0 - m_1}{m_0}$$

Where - m_0 is the mass in grams of the sample before reaction

m_1 is the mass, in grams, of the sample after reaction

The CSR is expressed as a percentage by mass, is given by the formula

$$CSR = 100 \times \frac{m_2}{m_1}$$

Where - m_2 is the mass, in grams, of the fraction of the sample >9,5 mm after tumbling

m_1 is the mass, in grams, of the sample after reaction

(British Standards Institute, 2018)

The aims and tolerances of these tests as agreed with the coke ovens are listed in the table below.

Measure	Min – Max	Target
M40	≤ 79 %	≤ 79 %
M10	≤ 8 %	≤ 8 %
Mean size		55 – 65
CSR	> 62 %	≥ 64 %
CRI	≥ 21 - ≤ 26 %	
Ash	≤ 10.5 %	≤ 11 %
Moisture	< 7 %	≤ 6 %

Figure 12 - Coke Quality Measures and Targets (Tatasteel, 2019)

Coke production

Coke quality and properties are dependent on the coals used to make the coke, and the coke making process. Therefore, an understanding of these processes would be beneficial. Coke is produced by the destructive distillation of coal, the coal is heated up to temperatures of around 1100°C in an oxygen deficient atmosphere, this drives off any volatiles present and prevents the coal from burning, leaving behind a carbon material called coke (Han, 1993). Morfa coke ovens were commissioned in 1981. It consists of two rows of ovens called batteries containing 42 ovens per battery. The ovens are 6 meters high, 16 meters long and 450mm wide with a 75mm taper. Each oven can hold 35 tonnes of coal and produces 25 tonnes of coke over an 18 hour period (METaL, 2016). There are two types of coke ovens used for the production of metallurgical coke for use in a blast furnace. They are the by-product coke oven and the non-recovery coke oven. Morfa, a by-product oven collects the off gas produced and an adjacent chemical plant removes any by-products such as crude benzole, naphthalene, tar and coke oven gas. These by-products are valuable and so can be sold. Coke oven gas has a high calorific value making it useful as a fuel in other steelmaking processes.

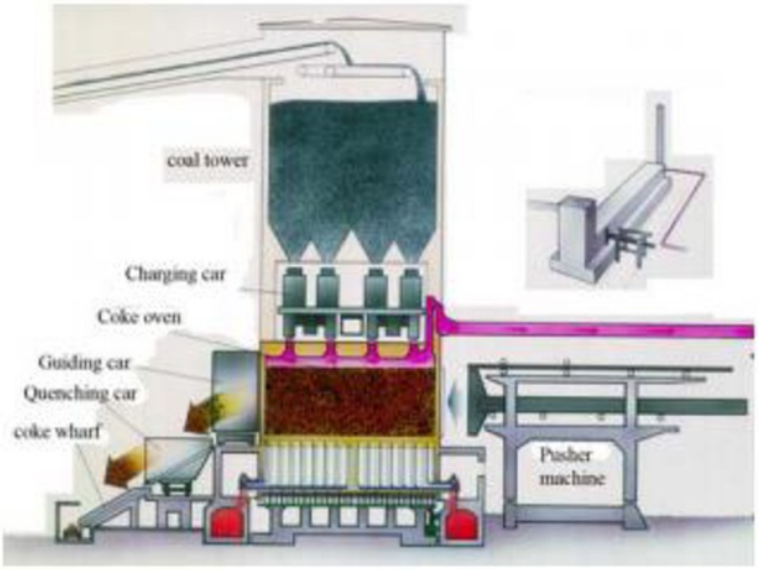


Figure 13 - Diagram of a typical by-product furnace (Hilding, 2005)

The difference with a non-recovery coke oven is that it does not collect any by-products as its name suggests, instead the coke oven gas is burnt off, and the surplus energy produced is used to produce steam for power generation (United States Environmental Protection Agency, 1995). Non-recovery coke ovens give off significantly less pollution, are smaller, cheaper to run and they are able to use cheaper coals to produce good quality coke but the cost of building them along with a power generating plant means they are only economical when emission requirements are strict and electricity / energy prices are high (Towsey, et al., 2010).

The transformation of coal to coke takes place as follows, heat from the oven walls heats the coal in contact with it very rapidly, at temperatures between 375°C - 475°C the thin layer of coal near the walls, softens and fuses becoming plastic. As temperatures raise to 475°C-600°C volatile gases are released followed by a decrease in plasticity and re-solidification of the coke occurs. Between 600°C - 1000°C the coke stabilises, shrinks, and more gases are evolved. During the plastic stage, the two plastic layers move from the oven walls, towards the centre where they meet. Liberated gases are trapped creating a build up of pressure which is transferred to the oven walls. Once the plastic layers meet the entire oven has been carbonised. The coke is now ready to be pushed from the oven before being quenched with water to prevent oxidisation of the coke (Loison, et al., 1989).

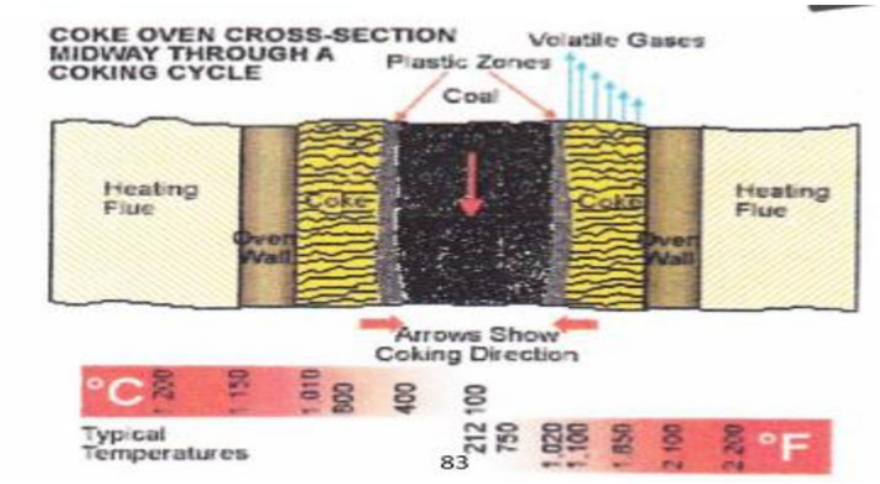


Figure 14 - Cross section through a coke oven during a coking cycle (METaL, 2016)

Coal selection and blending

There are many types of coal available from mines around the world and the classification depends on the age of the coal seam and the pressure it has been under. They are classified by a rank system, which is based on the degree of fixed carbon content as measured by the volatile test. They can be separated into non-coking and coking coals. There are up to 600 coal deposits available around the world of all coal ranks (METaL, 2016).

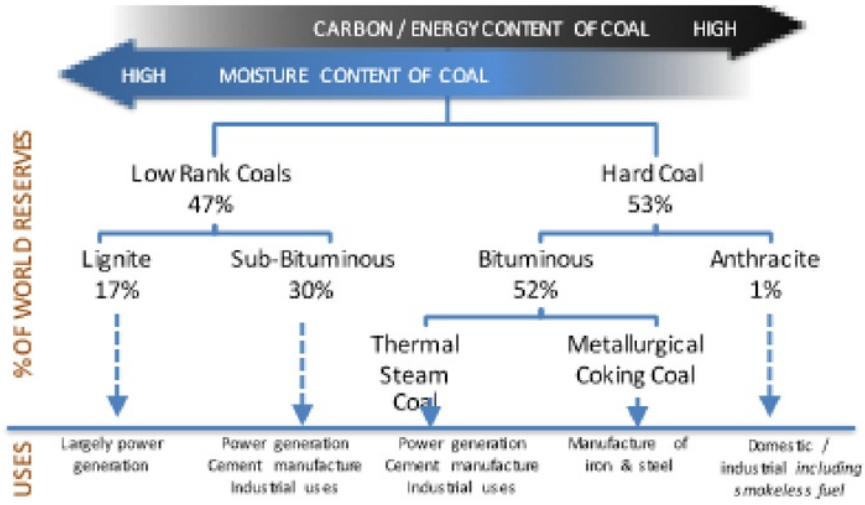


Figure 15 - Coal types and uses (METaL, 2016)

Non-coking coals are unsuitable for the steel industry as are many of the coking coals due to high levels of sulphur, phosphorus and alkali's which would contaminate the hot metal produced at the blast furnace. Some coking coals are also unsuitable due to poor physical properties such as high ash, high reactivity, low fluidity or low CSR in the coke produced. This only leaves around 50 - 60 coals worldwide suitable for coke making (Han, 1993) (Díez, et al., 2002).

Han 1993 explains that due to the availability and price of these coals it has become necessary to blend numerous coals with typically 6 – 8 different coals making up a blend. The objective of the blend is:

- Achieve the required coke quality, particularly strength (CSR)
- Avoid high coking pressure
- Reduce the need for expensive prime coking coals
- Control coke chemistry
- Control by-product yields

Representative sampling

Coke is analysed by the laboratories to assess its quality, this gives the coke ovens an indication how they are preforming and if its targets of coke quality are being met. Coke quality can be predicted as individual coals and blends of coals are tested on small scale ovens called pilot ovens. The cost of the coals can vary and so the coke ovens often change the coals used in the blend to try balance quality against cost. Although, as there are many variables inherent during the coke making process, these predictions are not always met. The analysis of the coke also allows the blast furnace to predict how the coke will behave inside the furnace. The main drivers for coke analysis are therefore process control, quality and cost.

To accurately assess the quality of the coke produced at Morfa coke ovens, a small sample should be obtained that represents the coke being produced during a 24 hour period. Considering that Morfa produces approximately 75 tons of coke every hour, a sample of approximately 180kg would be needed that represents 24 hours of coke making (~1800 tonnes). This 180kg then would need to be split down again for individual tests. A sample of 1 gram is needed for ash analysis for example. That 1 gram needs to be representative of the whole ~1800 tonnes.

The recommended method for sampling a material such as coke produced in this way is routine conveyor belt sampling during transportation of the coke to its final destination i.e. the

blast furnace (ISO, 2001). The international standards organisation (ISO) recommends that samples are taken using autosamplers from a falling stream. A falling stream sampler is situated at the end of a conveyor system as the material falls off the conveyor. A cutter moves through the material as it falls off the conveyor. The reason this method is preferred as it has less bias in the collection of the material than other methods.

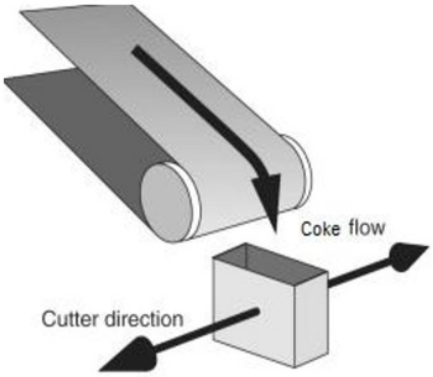


Figure 16 - Diagrammatic representation of falling stream sampling (Membrey, 2013)

A particulate material such as coke comes in many sizes and these particles can separate and settle and so it is important all the sample in a given area is taken. The standard recommends that the sample size should be three times the nominal top size of the coke. The nominal top size of Morfa coke is 100mm, therefore a 300mm sample should be taken with all the coke within that 300mm gathered leaving nothing behind, also the sampling tool should have no sample left on it (British standards institute, 2009). The falling stream method ensures all these conditions are met (Mortenson, 2015).

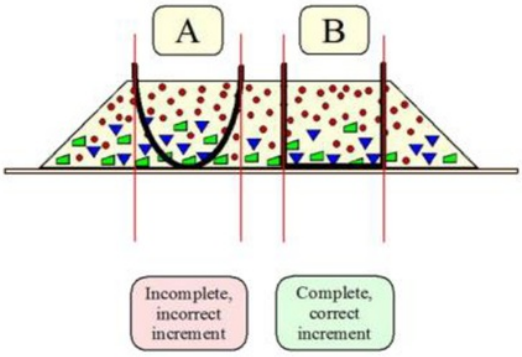


Figure 17 - The correct increment to take from a conveyor (Mortenson, 2015)

The new autosampler purchased by Tata steel and used for the collection of coke analysed for the purpose of this project is a type of cross belt autosampler called a hammer sampler. This autosampler has a cutter head with parallel side plates and a back plate, width determined by the nominal top size of the material. The cutter is swung perpendicular to the direction of the conveyor, cutting a swath of material, propelling it into a chute.

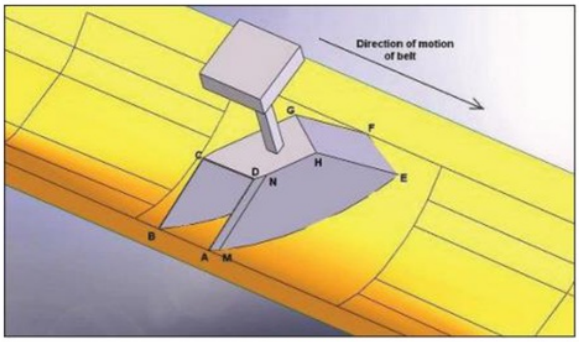


Figure 18 - The cutter on a 'hammer' cross belt autosampler (Robinson, et al., 2010)

The 'hammer' cross belt autosampler, although widely used, is not recommended by standards associations as there are a number of criticisms levelled against them. Namely, that all the material is not taken during a cut for a sample and some finer material can be left on the conveyor belt. Another criticism of this type of autosampler is that as the cutter moves at speed (~10m/s), it can cause breakage of the material. These issues could introduce bias into the testing of any material. The reason they are often used is due their relatively low cost and the ease in which they can be fitted to existing conveyor systems without any major changes to infrastructure. Retro fitting falling stream autosamplers would require changing the layout of the existing conveyor system and so would be significantly costlier (Lyman, et al., 2010). Although (Steinhaus & Minnitt, 2014) disagree with this opinion and attributes much of the criticism towards cross belt autosamplers to the inadequate design of early cross cut samplers and their poor integration into the plant. He argues cross cut samplers are more mechanically reliable and that there is little evidence of any significant difference in bias. That the choice of autosampler should be made after considering the type of material sampled and area of plant it is to be installed in.

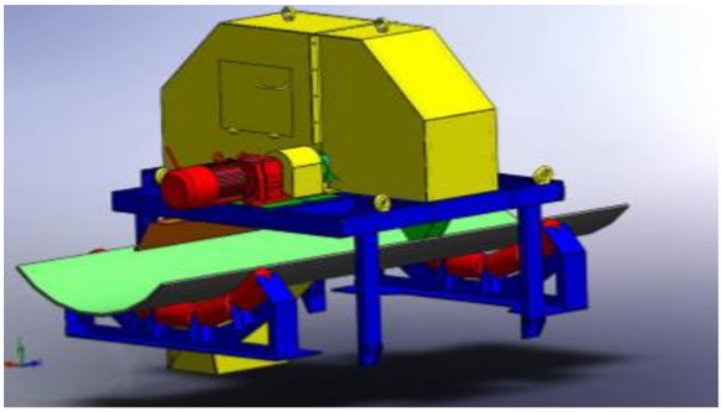


Figure 19 - Diagram of the autosampler purchased by Tata at Port Talbot steelworks (Wuxi YORK Corporation, 2018)

If there is no facility for mechanical sampling, then the ISO recommends manual stop belt sampling. Manual sampling involves stopping the conveyor belt and taking a full cross cut of the belt using a shovel. Care must be taken to remove all the sample off the belt and the cross section should be three times the nominal top size of the material (British standards institute, 2009).

The frequency of increments taken depends on the size of the bulk of material that needs testing, the material type and its particle size, the testing requirements, the required precision of results, expected bias, method of sampling and resource available to test. Once sampled the material must be kept dry and so should be kept in a watertight storage container. Once at the laboratory the sample needs to be mixed thoroughly and must be homogenous. The sample at this point could weigh hundreds of kilograms and will need to be spilt down into a manageable size, there a few ways of doing this and it depends on the material and particle size, again automation is recommended by ISO standards. Figure 20 shows an automated sample divider the sample can be poured in and it is spilt evenly into smaller parts by a rotating carousel figure 20 also shows a manual splitter called a riffler, the sample is poured in where it is split evenly in two, one half is kept the other is thrown away, this is repeated until the required sample size is achieved. By following these methods, a homogenous representative sample can be achieved (British standards institute, 2009)

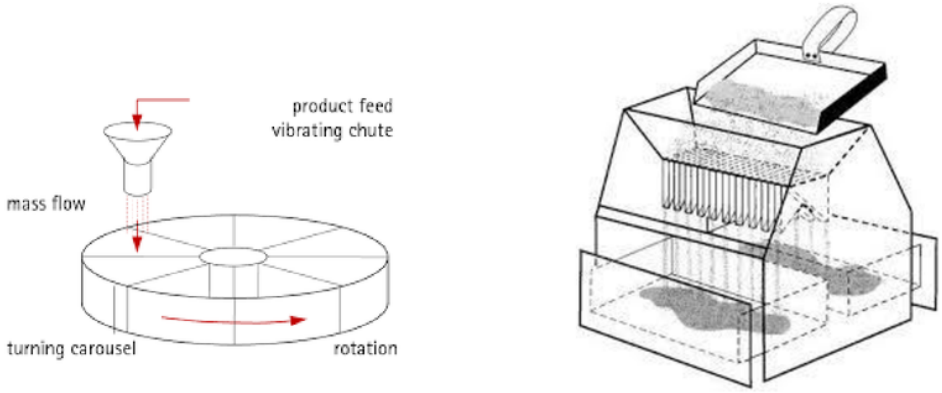


Figure 20 - Automated sample splitting & (Nenuwa & Adetiloye, 2016) Manual sample splitting with a riffler (Sympatec GbmH, 2018)

Next the sample would be prepared for analysis by methods such as crushing and pulverising. Preparation will depend on the analysis required.

Coke quality

Due to concerns from the blast furnace technical team, that the quality of the coke produced at Morfa coke ovens was contributing to blast furnace instability an investigation was initiated. Their opinion was that the variation seen in CSR results was too great. The agreement between the coke ovens and the blast furnace in SLA was a 64 aim and 62 minimum. After a run of results at or below 62 CSR it was decided that action was needed. The blast furnace technical team felt that coke with a CSR below 62 was a risk to furnace stability and so a decision was made to up the aim of the CSR to 67 with a minimum of 64. This was because on too many occasions results of below 62 were being observed and the variation in results was too great. A higher aim CSR would increase the quality of the coke and protect the furnace against the chance of producing low quality CSR coke.

Figure 21 below is a scatterplot graph showing the change in CSR aim. The graph also shows many results below the lower tolerance of 62% that triggered the coke quality investigation.

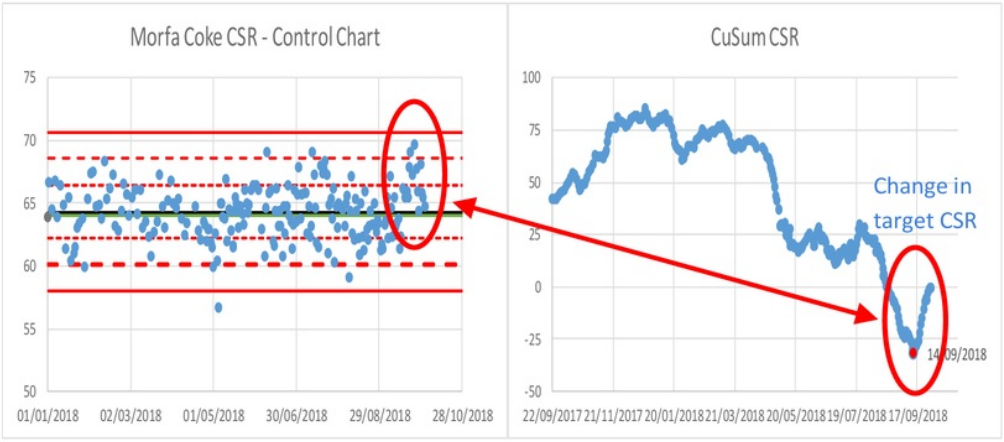


Figure 21 – Scatterplot and CuSum charts showing the drop in quality that triggered the change in CSR specification (Tatasteel, 2018)

To increase the aim of coke CSR to 67 the coke ovens would have to increase the amount of prime coking coal used in the coal blend, for every 1% raise in CSR it is estimated that this would cost £1.6 million per annum. The total cost to the business is estimated at approximately £8 million as not only more coking coal is required in the blend but opportunity is lost in using cheap local low or non-coking coals in the blend as these would be take out in the place of the prime coking coals. The below diagram shows the importance of coal selection to CSR.

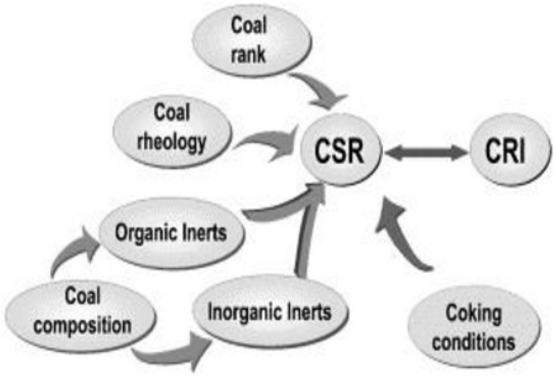


Figure 22 - Diagram showing the factors affecting CSR (Díez, et al., 2002)

The investigation focused on finding the possible sources of variation in coke CSR.

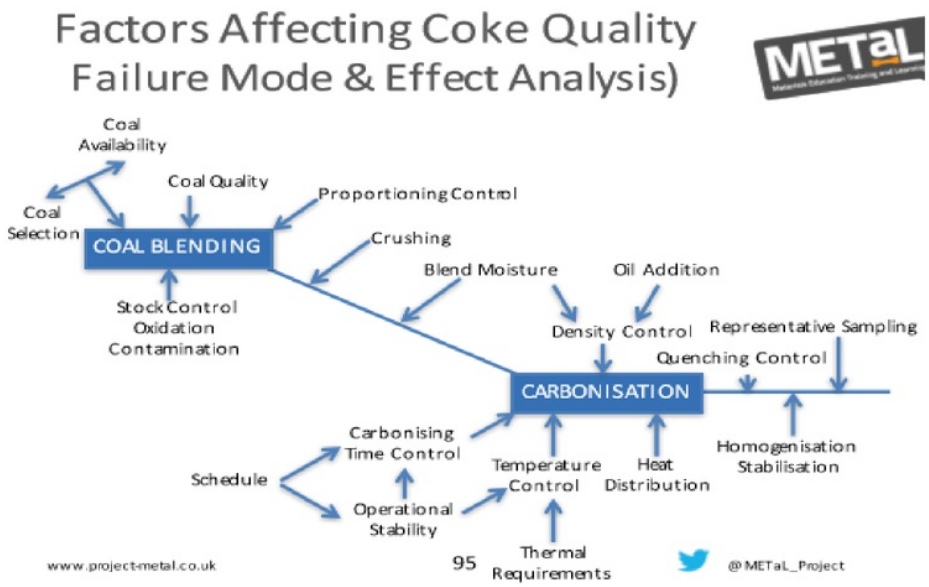


Figure 23 - Factors affecting coke quality (METaL, 2016)

The possible causes of variation were considered by the investigation team and it was decided that the most likely root causes of the variation in coke CSR, as shown in the fishbone diagram in figure 24, were:

1. Contamination in coal
2. Sampling, preparation and testing
3. Heating stability

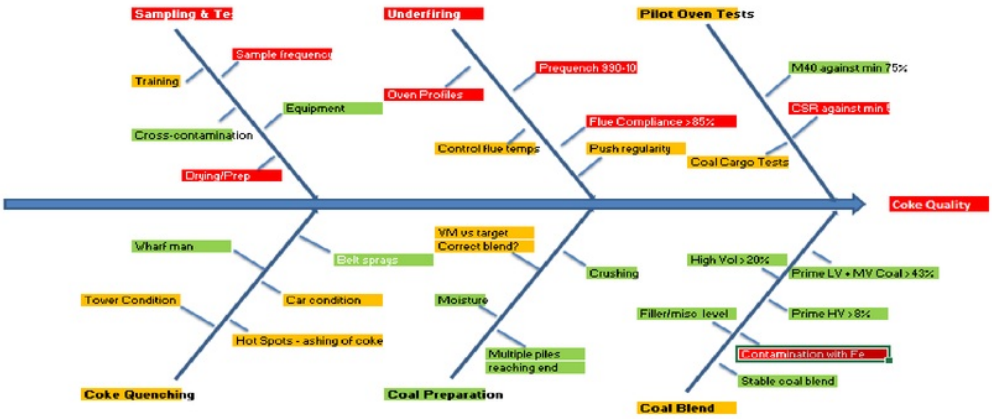


Figure 24 - Root cause analysis of coke quality variation carried out at coke ovens (Dickinson, 2018)

Once decided and implemented the coal blend does not often change, also the properties of the coals selected in the blend are known as they have been analysed at the laboratory.

Predictions can be made on the properties of the coke produced based on the coals selected (Álvarez, et al., 2007).

For every CSR result that fell below the set minimum 62 the coke sample was sent to the analytical laboratory for a chemical analysis, it was found that in some cases the low CSR result could be explained by a high iron content. Iron acts as a catalyst during gasification of coal it also causes organised crystalline structures within the iron/carbon interfaces leading to the creation of weak spots in the coke thereby reducing its strength (Li, et al., 2014) (Hilding, 2005).



Figure 25 - Iron contamination in coke sample (note orange spots) (Waters, 2018)

This iron contamination is a result of poor stockpile storage and reclamation management. Imported coal is discharged from the vessel at the harbour via conveyor belts the coal is stockpiled at the harbour stockyard before being reclaimed and transported to the coal stockyards via conveyor. The vessel discharge chutes also service iron ore cargoes and need to be thoroughly cleaned after use, if this is not done or if the chutes are not cleaned sufficiently then coal contamination can occur. The stockyards are arranged so that specific materials are designated areas of the yards, if the yards are overstocked or mismanaged coal could be offloaded in an area where an iron bearing material had been stored before it, when the coal is reclaimed the reclaimers pick up the iron bearing material along with the coal at ground level.

Heat stability at the coke ovens can also affect CSR. Heating stability depends on:

- Coke oven push regularity
- Stable fuel gas calorific value (CV)
- Consistent supply of fuel gas

Coal is turned to coke over a period of approximately 18 hours, the ovens are pushed in sequence with every 5th oven pushed approximately every 20 minutes. If there are issues such as coke getting stuck in the oven then the push regularity rate drops, the average temperature in the ovens rises and coke soak times increase. All these factors can have an adverse effect on coke quality.

The CV of the fuel gas is also related to coke oven temperatures and so quality, blast furnace gas has a lower CV than coke oven gas and it therefore it is enriched with coke oven gas to provide enough energy for the coke ovens. If the CV of the blast furnace drops then more coke oven gas is required, there is a delay before the drop in CV is registered and acted upon which causes a temporary drop in coke oven temperatures, see figure 26.

Both blast furnaces do no always operate together, if for whatever reason one furnace goes offline then at least one battery is required to switch to Blast furnace gas under firing as the COG is required at the power plant, this switch temporarily results in a drop of heat input into the ovens. The increased frequency of the switches has increased significantly over the last few years and is thought to contribute to CSR variability.

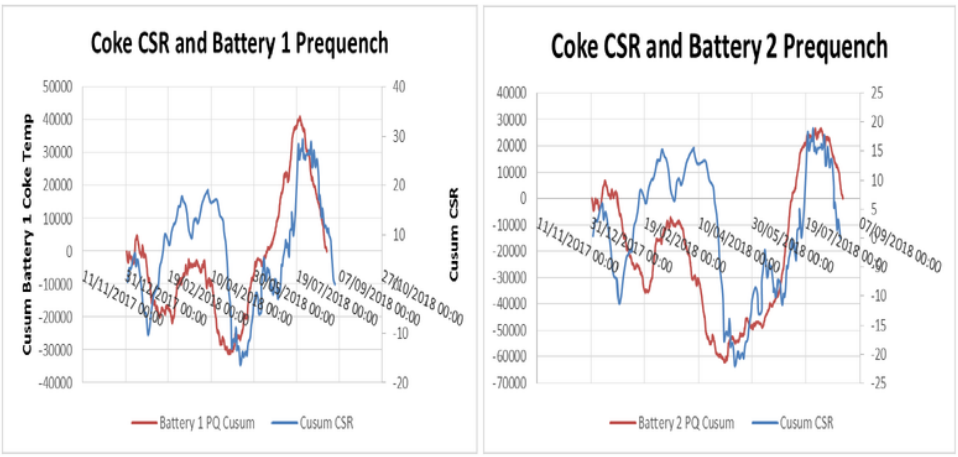


Figure 26 - CuSum graph showing correlation between coke temperatures and CSR (Dickinson, 2018)

Although these factors can explain some of the CSR results that fall below the minimum control limits, they do not explain the amount of variation in results observed.

Sampling and preparation of Morfa coke

Morfa coke is sampled by contactors working on the coke wharf at Morfa. Their main roles are to maintain the coke belts and keep them running. As they work in the vicinity of the sample point, in 2016 they were given responsibility for sampling coke. The collapse of the coke

bunkers and reduction in laboratory manning meant that a new sampling regime was required. The laboratories no longer worked nightshifts and a new sample point was needed. Coke was now going to be transported to the blast furnace via lorries. A stacker was hired and fitted to the end of the coke conveyor, the coke would be unloaded from the stacker into a stockpile. Front loaded shovel diggers would then load the lorries from the stockpile.



Figure 27 – Coke Stockpile after stacker at Morfa Coke Ovens (Waters, 2018)

It was agreed that for moisture, chemistry and CSR the sample would be taken from a point just before the coke gets to the stacker. The wharfman would stop the belt every four hours and take a full cross cut of the belt with a shovel into a watertight plastic barrel. The laboratory would then collect the coke twice a day at 07:00 and 17:00.



Figure 28 - The belt sample point on 1302 conveyor (Waters, 2018)

For the micum test, a larger sample is required (approx 50Kg) meaning the belt sample would not suffice. It was decided that every four hours the front-loaded shovel digger would take a load from the stockpile and drop a pile onto a concrete plinth (shown in figure 29), the laboratory technician would then take a cross cut from each pile and take it back for testing, again at 07:00 and 17:00.



Figure 29 - Piles of coke left by the front-loaded shovel digger at Morfa concrete plinth (Waters, 2018)

For CSR and micum testing the 07:00 and 17:00 samples are mixed together as a 24 hour composite.

Once back at the lab the CSR/CRI, moisture and chemistry sample contained in blue barrels is crushed in a jaw crusher, the jaws in the crusher are set so that between 10% - 30% of the material is crushed to 19mm – 22.4 mm. The sample is then thoroughly mixed and split by a method called cone and quarter, this involves putting all the material on the floor before taking shovelfuls of the material and creating a separate conical pile, the pile is flattened and then cut into quarters. The method is used to split bulk samples down to a manageable size. To mix, the quarters are shovelled back a small bit at a time back into a conical pile and the process repeated three or four times before being spilt again into quarters. At the end the four quarters are used, two quarters are taken for moisture analysis in duplicate, one quarter for chemistry and one quarter for CSR/CRI testing.

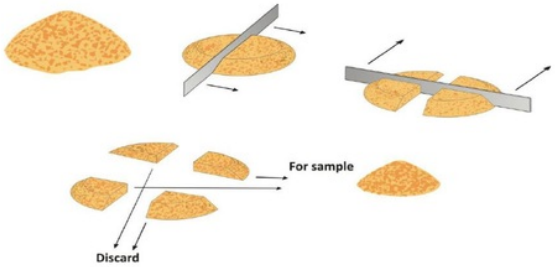


Figure 30 - The cone and quarter method (Alakangas, 2015)

Reducing variation introduced through sampling and preparation

Once the sampling and preparation of coke was identified as a probable source of variation in CSR by the coke ovens technical team, a meeting between the coke oven technical team, blast furnace technical team and the laboratories raw materials management was called. It was decided that the current methods of sampling and preparation needed to be changed. The sampling regime that was forced upon the business in 2016 was no longer acceptable. The coke ovens team were charged with improving heating stability and reducing coal contamination while the laboratory, specifically the author as raw materials technologist, was given a project to reduce variation in coke analysis introduced by sampling and preparation. To aid this project, a member of the blast technical team had sourced a comparatively low cost, cross belt autosampler of the hammer type and was to purchase it immediately.

All current sampling and testing practices were to be audited and reviewed to see if they were being carried out correctly and to evaluate if any improvements could be made. The arrival of the autosampler would also mean some new procedures and practices would need to be established.

Criticisms of the current sampling regime from 1302 conveyor (Figure 28) were as follows:

- The frequency of samples was lower than ISO standard recommendations.
- The sample point was not a designated sample point and sampling from this point using a shovel was awkward and difficult.
- A full cross of the belt was difficult to achieve.
- Recommended method of sampling was by autosampler (falling stream).
- Coke at this point is recently quenched and there are often large amounts of steam – visibility is poor.
- The wharfman taking the sample has other roles and sampling may not be his priority.
- The wharfman would not be trained in sampling techniques and would not be aware of representative sampling.
- Often, samples not available for collection – Not taken by wharfmen
- Number of conveyors stops recorded did not match number of samples expected – Wharfman not routinely collecting sample.
- Lab technicians were observed poorly mixing the samples.

Criticisms of the current sampling regime from the stockpile after the stacker (Figure 29) were as follows:

- The frequency of the sample was lower than ISO standard recommendations.
- Stockpile sampling not recommended – Size fraction segregation in stockpiles and poor sample representation.
- Front loaded shovel digger not recommended – Mechanical degradation of sample and poor sample representation.
- Risk of contamination from material on the floor.

These practices were open to random error, bias and variation.

Stockpile segregation is an issue with this form of sample collection. Stockpile segregation starts on the conveyor belt, the motion of the belt over the rollers causes finer material to settle at the bottom of the belt and courser material settles at the top figure 31, as the material is then discharged the particles at the top have a greater velocity than the particles at the bottom. The momentum of the course particles takes them towards the bottom of the stockpile and the finer material stays towards the top figure 32 (Nohl & Domnick, 2000).

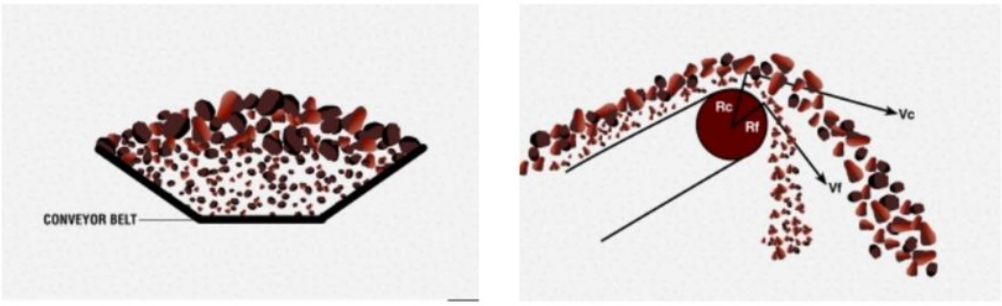


Figure 31 – Material segregation on a conveyor and conveyor discharge (Nohl & Domnick, 2000)

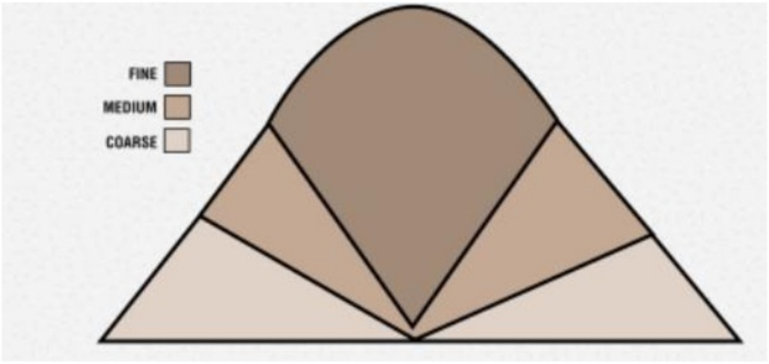


Figure 32 – Material segregation in a stockpile (Nohl & Domnick, 2000)

The installation of the autosampler would allow the laboratory to reduce or eliminate these influences.

The blast furnaces agreed that if the variation seen in CSR analysis could be reduced then the tolerance for CSR could be reduced back down to an aim of 64% with a 62% minimum. This would be reviewed in July 2019 to see the progress on the projects, once satisfied that the variation was in control then the limit would be reduced.

Implementation of the autosampler

The autosampler was sourced from China and was considerably cheaper than any alternatives from Europe, the Chinese autosampler cost approximately £10,000 while quotes from companies from around Europe came in at around £60,000. The autosampler was custom built to fit the coke stacker at Morfa coke ovens, as soon as it arrived on site, work started on installation. This took longer than expected due to some electrical issues and once it was operational further work would be required to make it fully automatic. Until then the autosampler would be used manually, the wharfman who was already responsible for taking the coke samples would be asked to press a button on a control box to take samples. There also was a need for some infrastructure to be built or purchased to deal with the sample once it was taken. As an interim measure until this was in place a plastic tube was attached to the autosampler to catch the coke and metal bins with lids would be used to store the coke.



Figure 33 – The Autosampler at Morfa coke ovens (Waters, 2018)

Some work would be required to validate the autosampler and make sure that its use did not impact the analysis of the coke in a negative way. Some change in the results are to be expected, it is hoped that the introduction of the autosampler would reduce variation in the results but there could be other changes. The action of the hammer, for example, is expected

to make a difference in the results. When taking the sample, the hammer will impact the coke as it takes it off the conveyor, the energy imparted from this impact could cause mechanical damage to the coke and so the results of the cold strength test, the micum, could change after introduction of the autosampler. Although as the coke would no longer be falling from the end of the stacker at a height of up to 10 meters it was theorised that this could negate the influence of the hammer. It was decided to run the old coke sampling method from the stockpile alongside the samples taken with the autosampler for two weeks and compare the micum results. It would be difficult to run all the tests alongside each other as there is not enough resource available in the laboratory and the coke ovens were keen to implement the autosampler as quickly as possible.

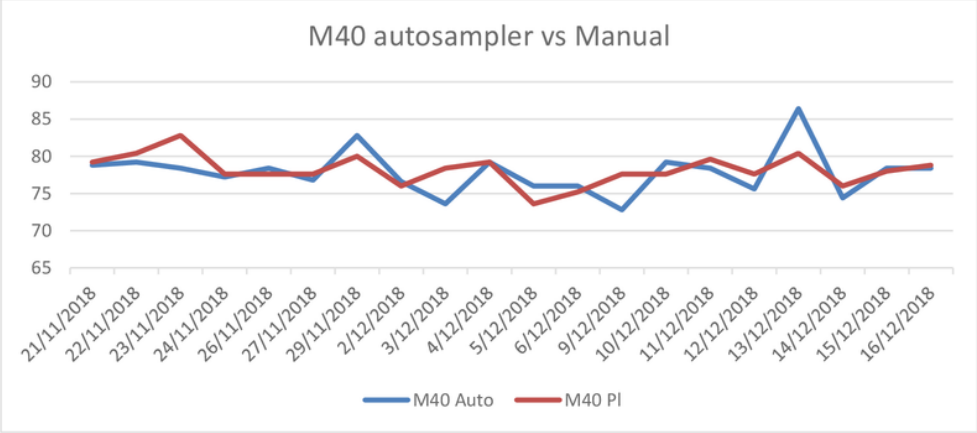


Figure 34 – Graph comparing the M40 of coke sampled from the autosampler versus coke sampled manually from the stockpile

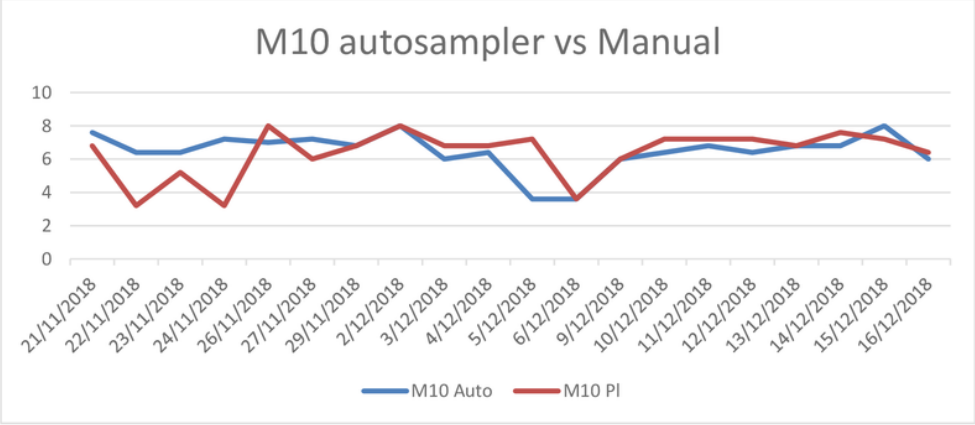


Figure 35 – Graph comparing the M10 of coke sampled from the autosampler versus coke sampled from the stockpile

	M40 Autosampler	M10 Autosampler	M40 Manual	M10 Manual
Mean	77.83	6.47	78.16	6.36

Figure 36 – Table showing the mean results for M40 and M10, autosampler versus stockpile comparison

From the comparison data there is no significant difference between the two sets of results, the means are very similar and therefore it can be said that the introduction of the autosampler will not introduce any new bias to the results. If any difference had been seen, then a new baseline for the M40 and M10 results would have had to be set and new control limits calculated.

It is expected that once the autosampler is fully operational that the amount of coke sampled would increase significantly. To be fully representative, a sample from every oven pushed would be required. The weight of a sample taken with the autosampler averaged 5Kg. An oven is pushed approximately every 20 minutes. That equates to 360 Kg of coke over a 24-hour period, with 2 collections a day that would mean the technicians would be required to move 180 Kg of coke from the coke ovens to the lab, mix the 180 Kg sample until homogenous and then split it down to a manageable size. The previous manual method involved the technicians collecting 60–80 Kgs per collection, a change in practices would be required.

One of the criticisms from the audit carried out on sampling and preparation of coke, was the mixing of the coke. Current practices involved splitting the sample in half into two bins and repeating three times. The sizing screens were removed from the bench and the sample was split over the middle partition into two bins. see figure 37.



Figure 37 - The micum screening bench with screens removed used for mixing coke samples (Waters, 2018)

Mixing is a vital part of sample preparation, if the sample is not made homogenous, then over representation of poor quality or good quality coke could occur in the sample and skew the results.

To cope with larger samples and make sure the sample is homogenous a new mixing procedure was implemented, four screens (Figure 38) are going to be removed and the sample split over four bins. This would be done four times and the sample bins would be changed around to further aid mixing. Two bins are kept for the micum test and particle size distribution composite. The second two bins are spilt again, one bin is thrown away as it is not needed, the coke in the second bin is crushed and then cone and quartered, the quarters are then used for moisture, chemistry and CSR/CRI analysis. The flow diagram in figure 38 helps explain the process. This method is the closest approximation to the ISO standard recommendations that is possible with the resources available to the laboratories. It is based on the riffle technique as shown in figure 20. Mixing coke is difficult due to its large particle size, instruments such as rifflers require their openings to be 2.5 times the nominal top size of the material used. Although there may be such rifflers available they are uncommon and often custom made. Ideally automation of the mixing process should be implemented as this would reduce variation caused by human error.

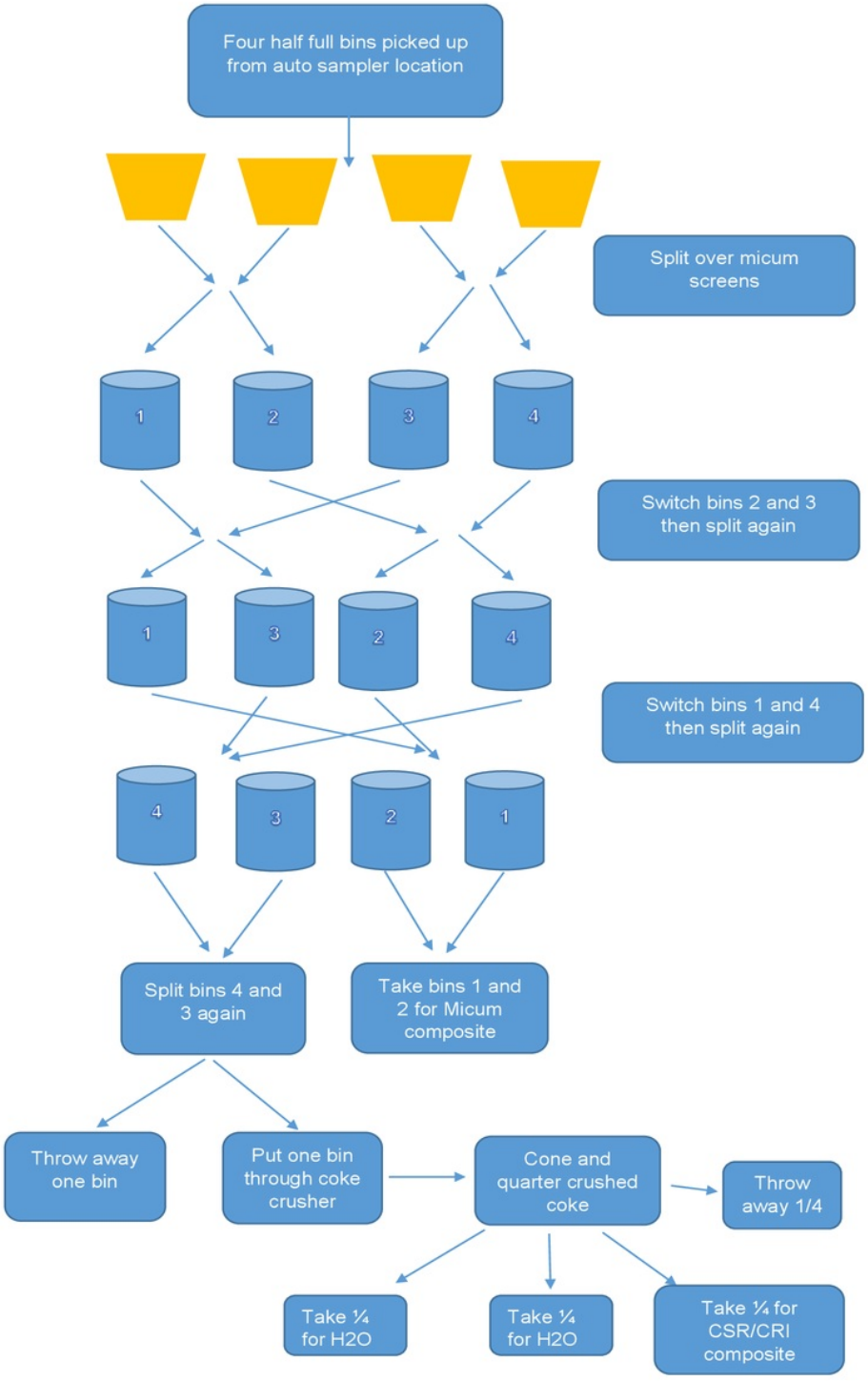


Figure 38 - The coke mixing process implemented for use after the autosampler installation (Waters, 2018)

Meetings were held between the coke ovens and laboratory to discuss implementation of the autosampler. Sample frequency was discussed as was handling and storage of the sample. The ideal sample frequency would be every 20 minutes but currently there was nothing in place to store the ~180Kg of coke that would accumulate over 12 hours. Also, the laboratory did not have the resource to increase the frequency of sample collection or the resource to transport ~180 Kg back to the sample preparation building. It was hoped that equipment to store and mix the sample would be in place before using the autosampler but it would take a few months to build and it had not yet been approved by the coke oven management and so it was decided that the autosampler was to be put into operation for sample collection as soon as it was ready, the laboratory had procedures in place and the technicians trained in these procedures.

The frequency of sample collection would have to remain the same, the wharfman would press a button to manually take a sample every four hours and the laboratory technicians would collect at 07:00 and 17:00. This new sampling regime would be very similar to the old one with the only difference being the autosampler would be taking the coke from the conveyor. Taking a sample every four hours will not give a full representation of the quality of the coke being produced, there are 88 ovens at Morfa and one oven is pushed every 20 minutes, taking a sample every four hours means only approximately 8.3% of the ovens are represented in the final sample. Each oven will produce coke of varying quality, depending on its condition and process parameters. Taking a sample every four hours increases the likelihood that a particularly poor or good oven is over represented in the sample.

To switch to full automation and increase sample frequency it was agreed that laboratory resource would have to be reviewed or a facility be put in place at the autosampler site to store and mix the sample at source.

Statistical Process Control (SPC)

SPC is a quality management tool that uses data and control charts and graphs to monitor a process to identify if it is functioning as it should. The process is measured over time and if there is unusual or undesirable variation in the process that can cause defects or issues, then the process can be corrected. Process control involves taking periodic samples from the process and measuring or testing them, the results are then plotted on a chart. Control limits are set and any deviation from these control limits will require investigating as the process can be said to be out of control. By monitoring these charts, poor quality can be detected early and defects prevented, as trends towards the control limits can be spotted and then corrected (Russel & Taylor III, 2011). All processes contain inherent variability and so everything

produced by the process will be different. Variables are derived from five sources, Machine, man, materials, environment and method. There are two types of variation seen in a process, the first is called 'common cause', this type of random variation is attributed to;

- Machine – Will never produce an identical product twice
- Man – Perform tasks slightly differently each time
- Materials – Can wear over time or have varying physical properties
- Environment – Temperature can cause contraction or expansion of parts
- Methods/ System – The sequence in which something is done can have an effect

These types of variation can never be eliminated although they can be reduced and represent the capability of a process. Common cause variation is predictable and if they were the only source of variation in a process, it would be stable and in control (Slack, et al., 2013). The other type of variation is called 'special cause', these are non-random and identifiable and if not corrected will cause poor quality, they are attributed to

- Machine – Not set up correctly or defective
- Man - Not properly trained or not following procedure
- Materials – Contains a defect or be unsuitable for its task
- Environment – Water can cause corrosion
- Methods/ System – The set up could be incorrect

Points out of specification in the control chart are likely to be special causes and require investigation. The control limits are set by measuring the amount of natural common cause variation in the process (Baradaran & Dashtbani, 2014). This is measured by first calculating the standard variation, the formula for standard variation is:

$$\sigma = \sqrt{\frac{\sum(x - \bar{x})^2}{n}}$$

Where

σ= standard deviation

Σ= Sum of

x= each value in the data set

$\bar{x}$= mean of all the values in the data set

n= number of values in the data set

A number of samples should be taken over time. A large sample size is preferable. The control limits are then calculated by

$$Upper\ control\ limit = mean + 3\sigma$$

$$Lower\ control\ limit = mean - 3\sigma$$

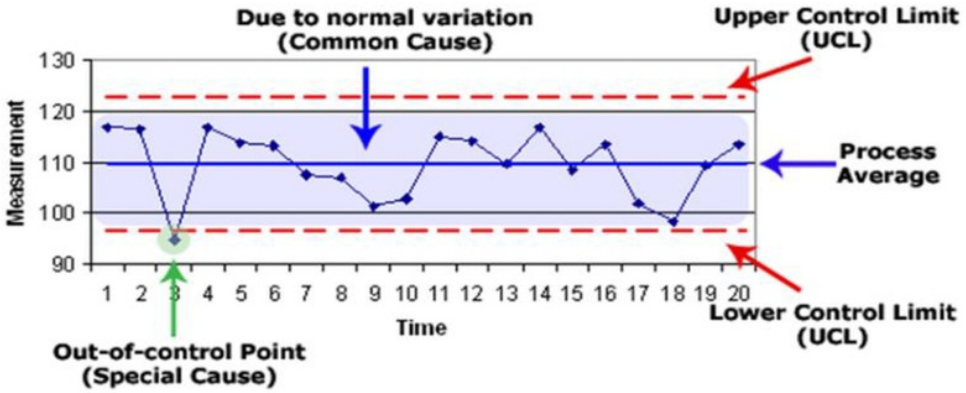


Figure 39 – A typical statistical control chart (Baradaran & Dashtbani, 2014)

(Russel & Taylor III, 2011) state that if the data points in the chart are counted by their frequency, then a normal distribution graph can be drawn (not all processes will be normal and the curve may be skewed). In a normal distribution curve +/- 1 standard deviation cover 68.3% of the data in this range, +/- 2 standard deviations cover 95.5% and +/- 3 standard deviations cover 99.7% as shown in figure 40 below.

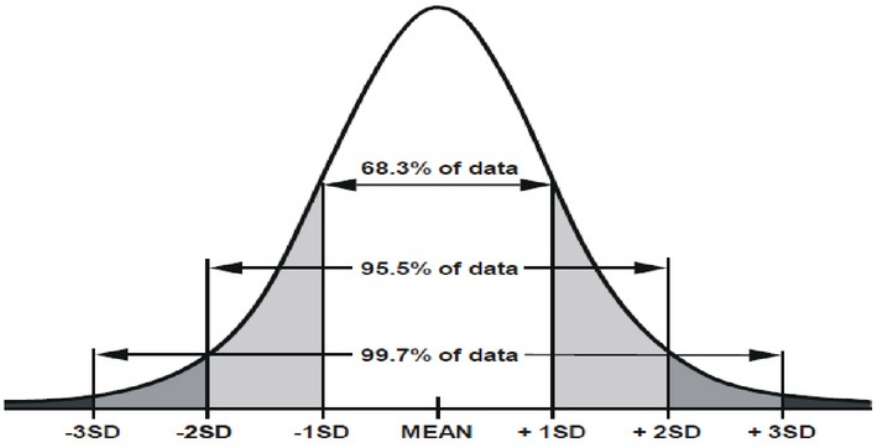


Figure 40 – Standard variation in a normal distribution curve (Jin, 2018)

Therefore, under normal operating conditions with only common cause variation present, the measurements of samples from the process should stay within 3x standard variation, 99.7% of the time. Any results outside of this is likely to be a special cause and therefore preventable (Russel & Taylor III, 2011).

If the standard deviation is small its means the data is clustered around the mean and there is little variation, the normal distribution curve is tall and narrow, a high standard deviation means the data is spread out and the normal distribution curve is short and wide, there is a large amount of variation (Slack, et al., 2013).

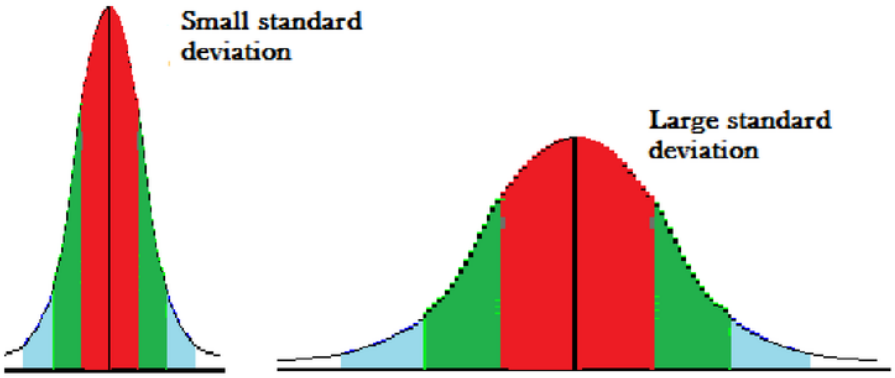


Figure 41 - Standard deviation in a normal distribution curve (Statistics How To, 2019)

Statistical analysis of coke quality analysis before installation of the autosampler

As there are other projects ongoing to reduce variation in CSR, a range of coke quality analysis, before and after the autosampler is installed is going to be compared. Comparison of just CSR and CRI would not enough to demonstrate that the autosampler had affected variation as it is likely the other ongoing projects will have their own effects. Not all coke quality analysis will be affected by these projects, iron contamination will have little if any influence on coke moisture results for example. It is hoped by comparing several coke quality analysis results that a reduction in variation through use of the autosampler can be demonstrated. The data has been displayed as control charts, with the measured analysis result on the y axis and time on the x axis. The control limits on some of the tests are not set as a standard SPC chart would be (by multiplying the mean result by 3x standard deviation) although these will be shown. The limits are set in the Coke ovens and Blast furnace SLA. The data used for

comparison before the autosampler was implemented, has been collated from between 1st October 2018 and 31st December 2018.

CSR/CRI

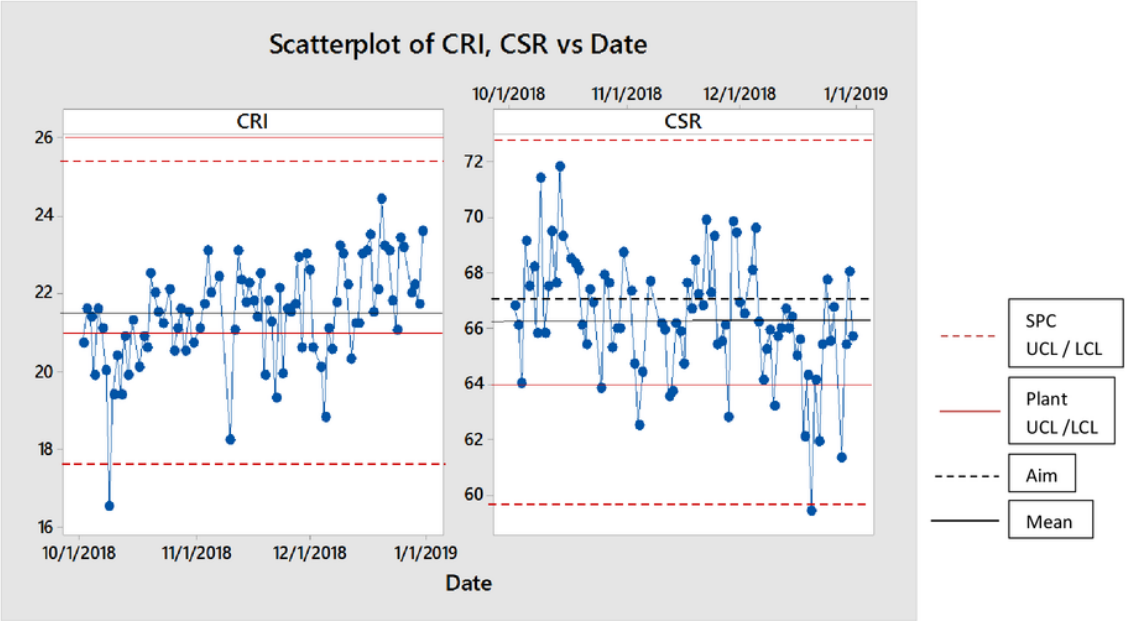


Figure 42 – SPC control charts for CSR and CRI between October 2018 and January 2019

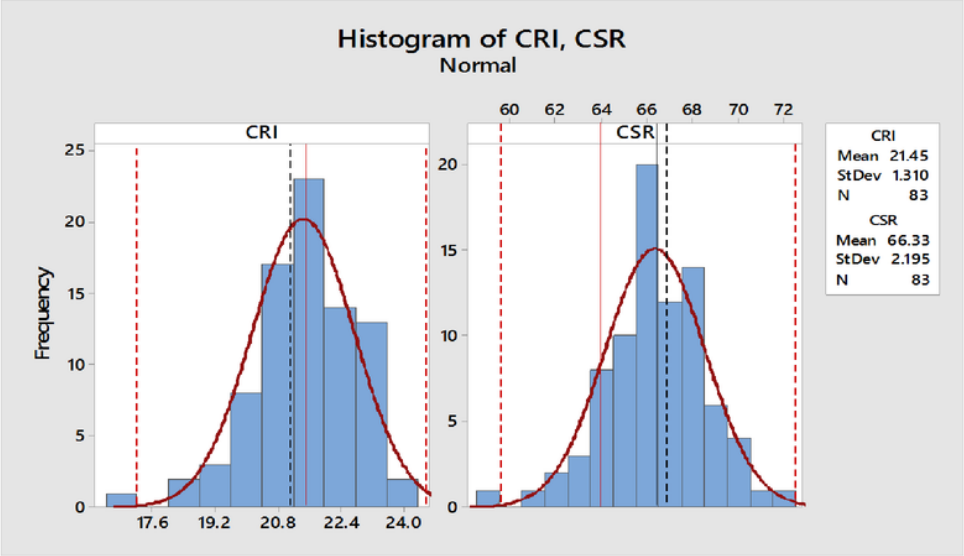


Figure 43 – Histogram and normal distribution of CSR and CRI results between October 2018 and January 2019

CRI analysis										
Mean	Standard Deviation	Min Value	Max Value	Range	Aim	Upper SPC control limit	Lower SPC Control limit	Upper plant control limit	Lower plant control limit	Percentage of results out of plant tolerance
21.45	1.31	16.5	24.4	7.9		25.38	17.54	≥ 21	≤ 26	36.51%

CSR analysis										
Mean	Standard Deviation	Min Value	Max Value	Range	Aim	Upper SPC control limit	Lower SPC Control limit	Upper plant control limit	Lower plant control limit	Percentage of results out of plant tolerance
66.3	2.195	59.4	71.8	12.4	67	72.89	59.72		64.00	12.05%

Figure 44 – Statistical tables of the CSR and CRI results between October 2018 and January 2019

CSR

The only control limit for CSR is a lower control limit, the blast furnaces and coke ovens set this limit in an SLA. The coke ovens to try not to produce coke with a CSR of less than 64% as they believe it would negatively impact the furnace. The labs have a CSR standard for monitoring the capability of the testing equipment and operators. The standard is made from a batch of Morfa coke, made homogenous and crushed and sized to the correct proportions for the CSR test. After being tested 80 times the standard variation on the standard was calculated at 1.6. This is the inherent common cause variation in the testing process. Two times this standard variation is 3.2 and gives 95% confidence that any test result will fall to within +/- 3.2 of the average/mean result. Although this does not take into account the variation inherent in the coke making process Therefore, to be confident of not getting a CSR of less than 64 the coke ovens must aim for a CSR of at least 67.2%.

The data shows that for the period between October and January 2018 that the mean CSR result was 66.3%, a little below the figure that the coke ovens would need to aim for to achieve a minimum of 64%. Using 3x standard variation upper and lower control limits, the process looks to be mostly in control with only one result out of specification but considering the 64- minimum set by the blast furnaces, then 12.05% of results fall below that minimum and are likely to be special causes such as poor heating, iron contamination or poor sampling. There also seems to be a slight downward trend in the results.

It is hoped that the introduction of the autosampler will reduce the standard variation by eliminating variation introduced through poor sampling methods and human error.

CRI

The blast furnace/ coke ovens SLA again set the limit for CRI, any result of over 26% would mean the coke is too reactive and it would over oxidise in the blast furnace and a higher coke rate would be needed. A low reactivity is also undesirable as this could lead to a build-up of coke in the lowest regions of the furnace (Geerdes, et al., 2009). An aim of ≥ 21 is desirable.

The laboratory standard has a standard deviation of 0.95, showing the inherent variation in CRI testing.

The data collected gives a mean of 21.45, close to the desired aim and the standard variation is 1.31 not a much larger than the standard deviation inherent in the testing. There was only one result below the lower SPC control level and none above the upper SPC control level suggesting the process is mostly in control and there were no results above the tolerance set by the customer.

If the autosampler reduces variation in CSR then as they are from the same test and the results have some inversely proportional correlation, then some reduction in variation should also be seen in the CRI results.

Moisture

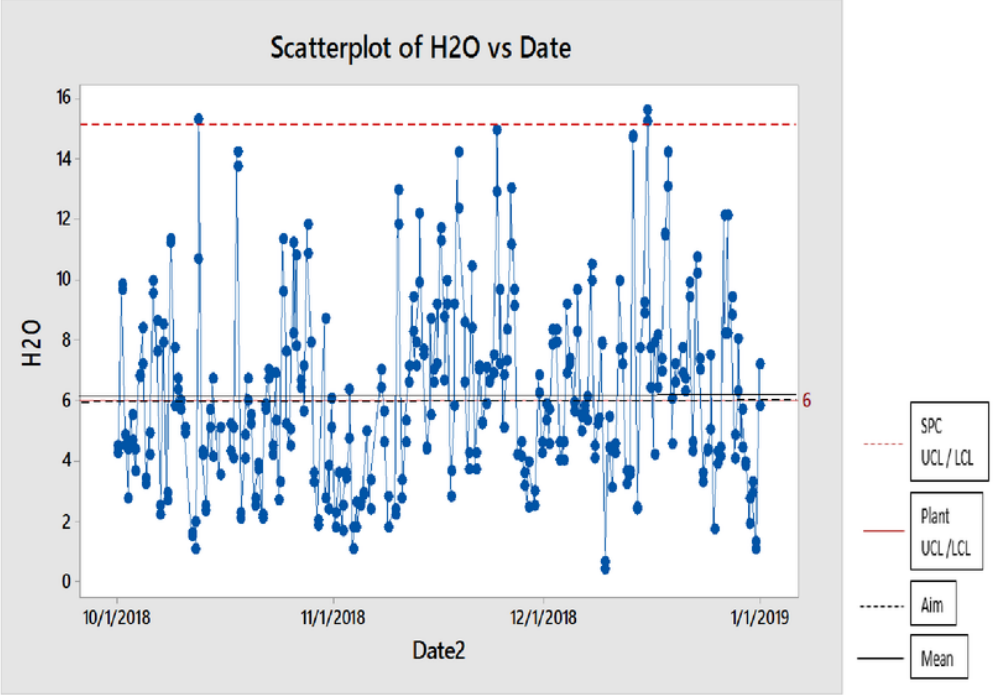


Figure 45 – SPC control chart of the moisture results between October 2018 and January 2019

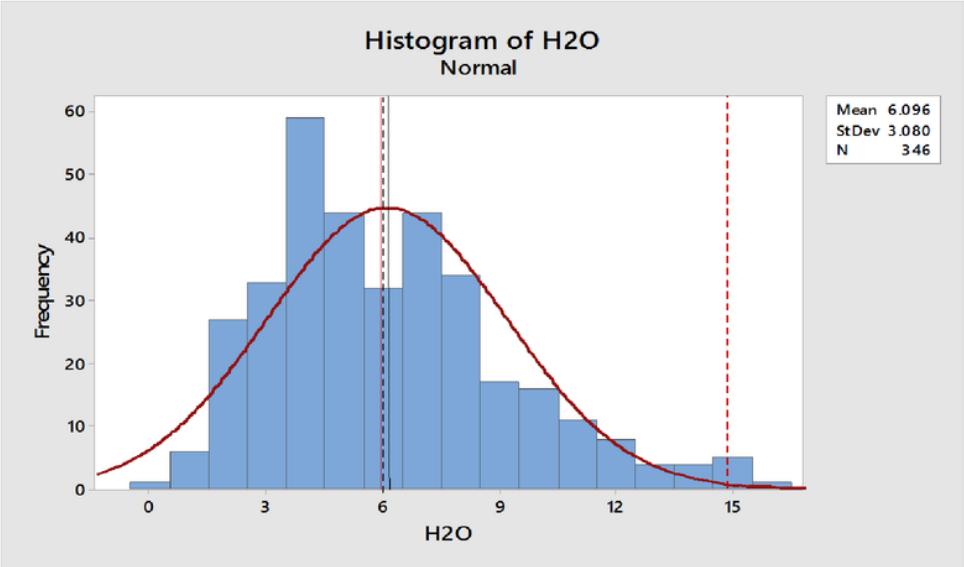


Figure 46 – Histogram and normal distribution of the moisture results between October 2018 and January 2019

Moisture analysis										
Mean	Standard Deviation	Min Value	Max Value	Range	Aim	Upper SPC control limit	Lower SPC Control limit	Upper plant control limit	Lower plant control limit	Percentage of results out of plant tolerance
6.10	3.08	0.40	15.58	15.18	< 6	15.34	0		6	55.66%

Figure 47 – Statistical table of the moisture results between October 2018 and January 2019

There is only a lower limit set for moisture analysis at < 6, anything higher is undesirable for the blast furnace as the energy consumed driving the moisture away is wasted and a higher coke rate is required to compensate the lost mass (Geerdes, et al., 2009). The mean result during this period is 6.1, just above the lower limit set by the plant. The standard variation is 3.08. There are a few results outside of the SPC upper limit indicating that the process is out of control, there is no lower SPC limit as 3x the standard variation falls below zero. 55.66% of the results fell above the 6% plant limit which is far from an acceptable amount.

There are many variables in the quenching process and quenching times being one, under filled ovens due to oven heating problems also cause issues as the quenching times are designed for full ovens (METaL, 2016). Quenching practices need to be improved, better control is needed, however it is predicted that use of the autosampler for moisture analysis could the reduce variation as it is theorised that as the coke is steaming after being quenched, visibility at the sampling point might be poor, this may deter the wharfman from taking a sample during this time. If this is the case, over representation of overly dry and overly wet coke could be in the data.

ASH

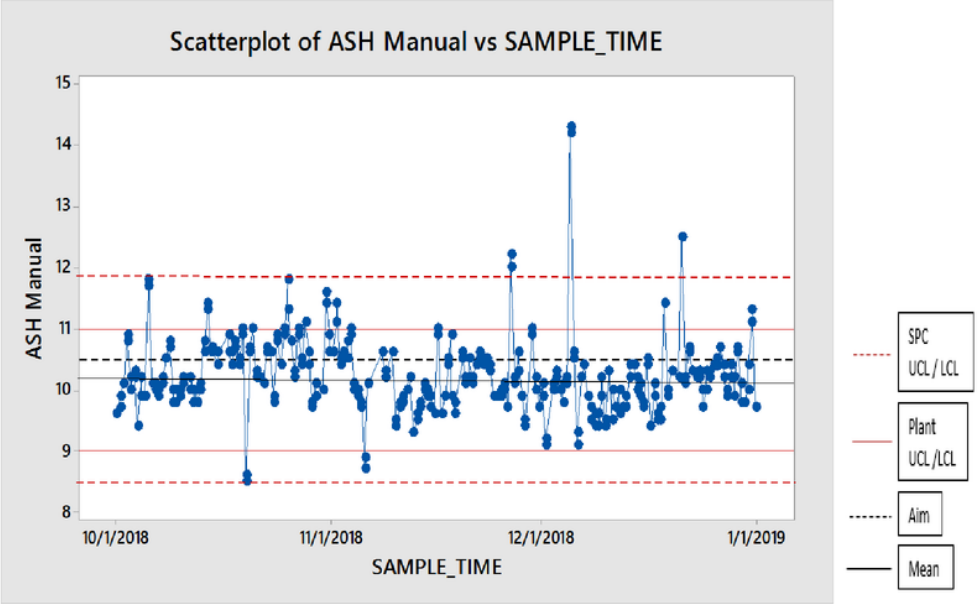


Figure 48 – SPC control chart of the ash results between October 2018 and January 2019

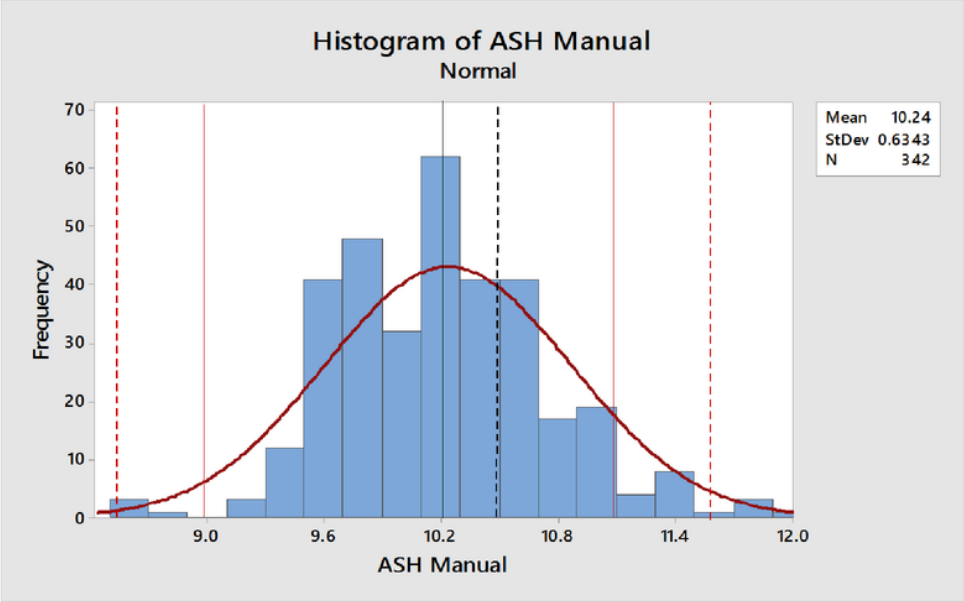


Figure 49 – Histogram and normal distribution of the ash results between October 2018 and January 2019

Ash analysis										
Mean	Standard Deviation	Min Value	Max Value	Range	Aim	Upper SPC control limit	Lower SPC Control limit	Upper plant control limit	Lower plant control limit	Percentage of results out of plant tolerance
10.24	0.63	8.50	14.30	5.80	≤10.5	11.87	8.54	11	9	9.31%

Figure 50 – Statistical table of the ash results between October 2018 and January 2019

The mean and the aim are close together and the standard variation is 0.63, however there are many results over the upper SPC limit suggesting that the process is out of control. There are also numerous results outside of the limits set by plant, 9.31% alltogether. The plant limits and the SPC limits are relatively close suggesting that the plant limits were set using SPC and either the process was more in control, a 2x standard deviation limit was set or the plant decided on stricter limits than those derived from the SPC charts. Like the CSR, some of the high ash analysis results can be atributed to Fe contamination, which can be confirmed by chemical analysis of the ash. There is corralation between low CSR results and high ash results (Álvarez, et al., 2007).

It is anticipated that there will be little effect on the ash analysis after the autosampler is introduced as the ash result is mostly dependant on the composition of the coal blend (Nag, et al., 2009). Although, under coked coke will have an effect on ash analysis, so a small reduction in variation is possible if the frequncy of under coked coke over represented in the sample is reduced.

Mean size

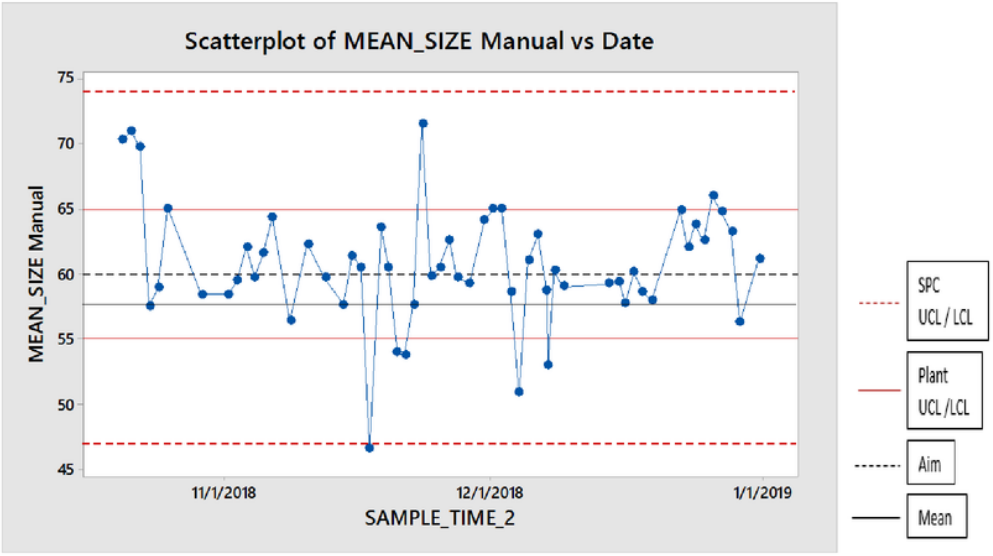


Figure 51 – SPC control chart of the mean size results between October 2018 and January 2019

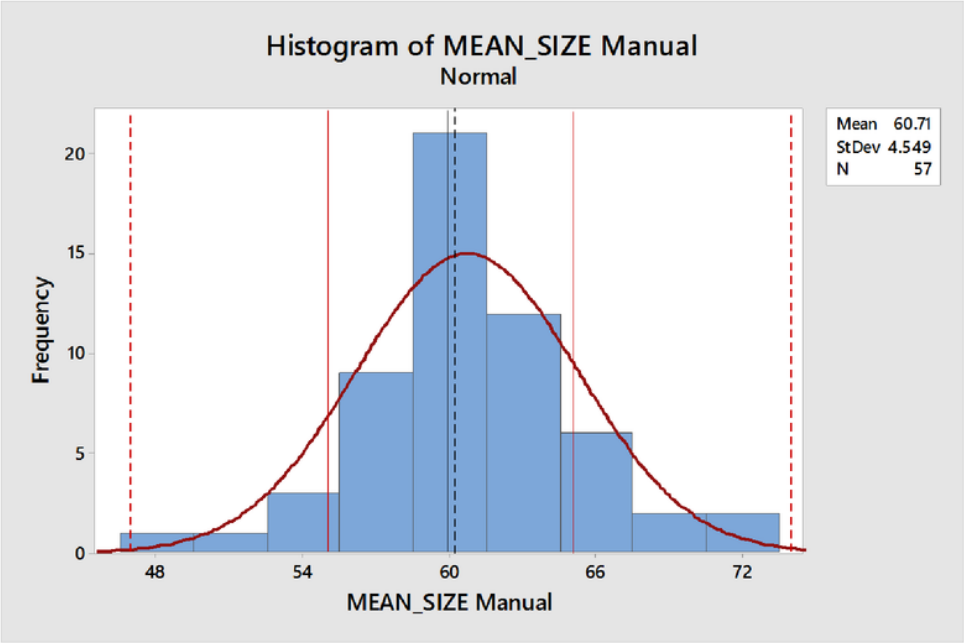


Figure 52 – Histogram and normal distribution of the mean size results between October 2018 and January 2019

Mean size analysis										
Mean	Standard Deviation	Min Value	Max Value	Range	Aim	Upper SPC control limit	Lower SPC Control limit	Upper plant control limit	Lower plant control limit	Percentage of results out of plant tolerance
60.71	4.55	46.54	71.57	25.03	60	74.36	47.06	65	55	27.76%

Figure 53 – Statistical table of the mean size results between October 2018 and January 2019

The mean of the mean particle size is 60.71 and the aim is 60, these results are close but there is considerable deviation from this mean at 4.55, indicating there is a large spread between the lowest and highest results (25.03) this may be the capability of the process but there may be scope to lower it. There is one result outside of the SPC limits and the process could be said to be mostly in control, alternatively 27.76% of the results are outside of the operational limits.

As already discussed in the report taking samples from a stockpile is not recommended, as the material will segragate in the pile, with larger material settling at the bottom and finer material settling towards the top. It is hoped that by using the autosampler to take samples that a reduction in variation will be seen, as it is unknown from where in the pile the different digger drivers are taking the samples. By removing these types of bias, the autosampler coke sample should be more representative of the actual mean particle size of the coke produced.

Micum M40 and M10

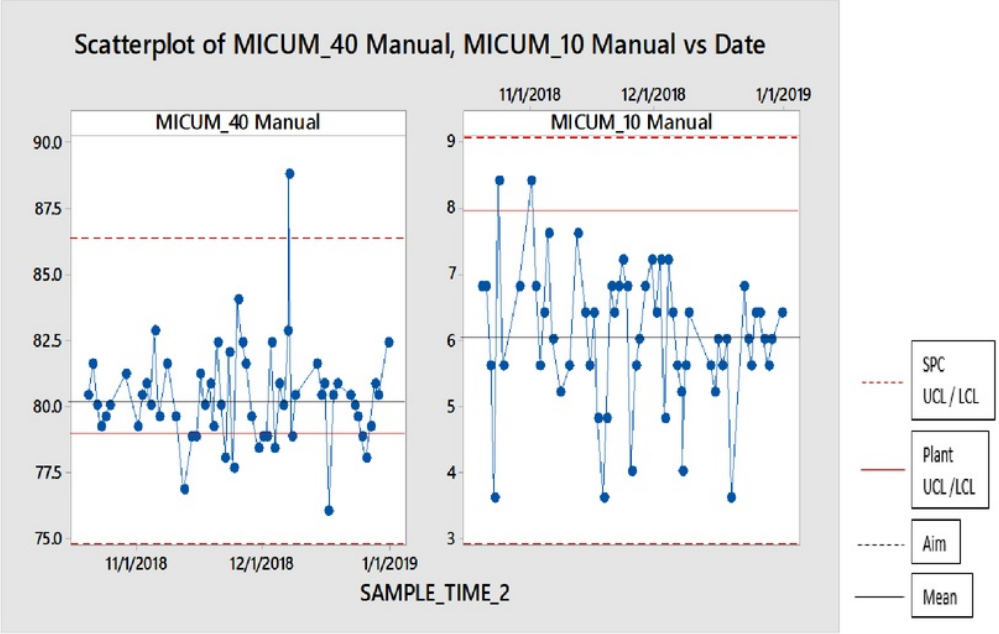


Figure 54 – SPC control chart of the M40 and M10 results between October 2018 and January 2019

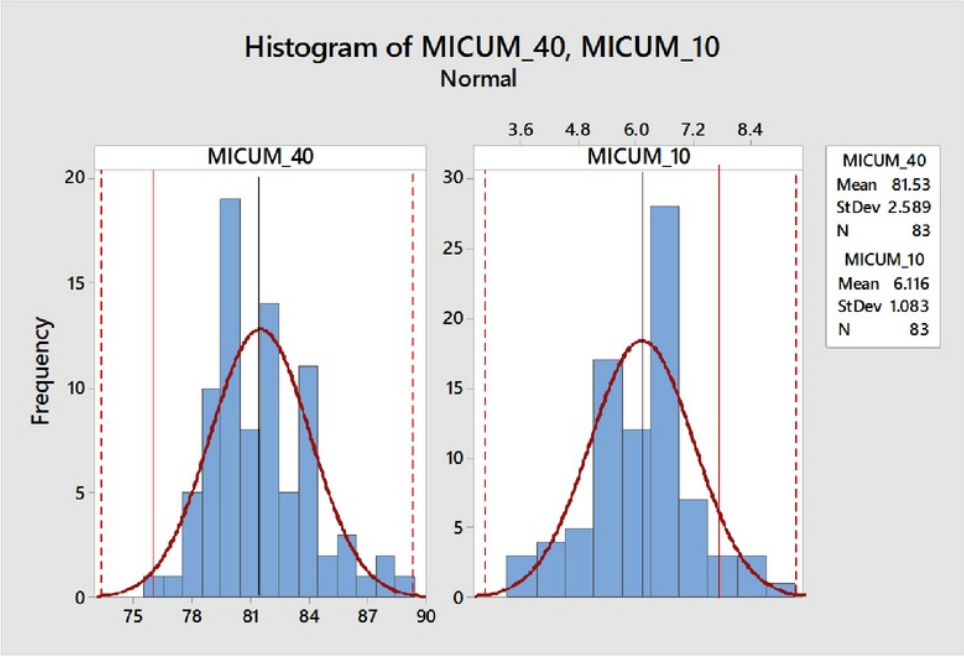


Figure 55 – Histogram and normal distribution of the M40 and M10 results between October 2018 and January 2019

M40 analysis										
Mean	Standard Deviation	Min Value	Max Value	Range	Aim	Upper SPC control limit	Lower SPC Control limit	Upper plant control limit	Lower plant control limit	Percentage of results out of plant tolerance
80.3	1.92	76	88.8	12.8	≥ 79	86.06	74.54		≥ 79	24.62%

M10 analysis										
Mean	Standard Deviation	Min Value	Max Value	Range	Aim	Upper SPC control limit	Lower SPC Control limit	Upper plant control limit	Lower plant control limit	Percentage of results out of plant tolerance
6.12	1.08	3.60	8.80	5.20	> 8	9.37	2.87	8.00		4.09%

Figure 56 – Statistical tables of the M40 and M10 results between October 2018 and January 2019

M40

The limit set by the blast furnace/coke oven SLA for M40 is <79. The mean for the M40 was 80.3 which is above the minimum control limit, although 26.62% of results are outside of the agreed plant control limits. There is only one result out of specification for the SPC and plant control limits and so from an SPC point of view the process can be regarded in control for the majority of the time.

The sampling method, from a stockpile, is poor because of particle size segregation. Due to this, there may be a difference in standard deviation between the two sampling methods. The initial particle size may have an effect on the M40 result. If there is a high proportion of larger material in the sample, then there is a higher chance of having +40mm coke after tumbling.

M10

The aim set by the blast furnaces/ coke ovens SLA for M10 is >8. The percentage of results outside this control tolerance is 4.09% The mean of the M10 results was 6.12 with a standard deviation of 1.08. There are no results outside of the SPC tolerances indicating the process is in control.

Again, due to the particle size segregation, switching to autosampler may have an effect on the results for M10.

Statistical analysis of coke quality analysis after installation of the autosampler

It was originally intended that three months before and after implementation of the autosampler would be compared. However due to delays during installation and to allow enough time to analyse the data and write up the project, it was decided that two months of data collection would have to suffice. The autosampler was implemented on February 7th 2019 and data was collected until 28th March 2019, after this date the autosampler experienced electrical issues and was not used for a week so it became necessary to stop collecting data.

CSR

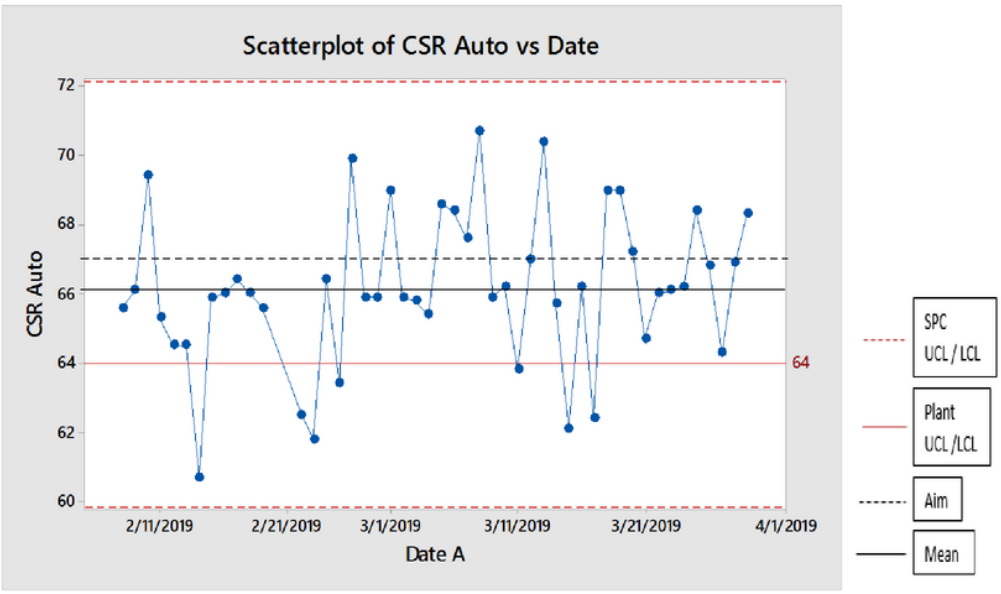


Figure 57 – SPC control chart of the CSR results from coke sampled from the autosampler

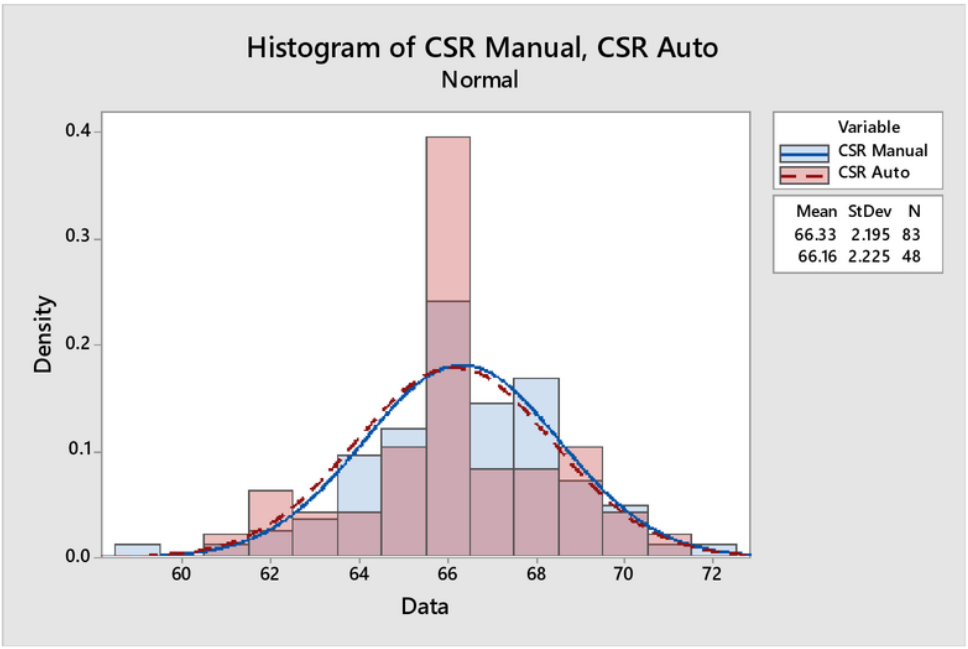


Figure 58 – Histogram and normal distribution comparison of CSR results sampled manuallly and with the autosampler

CSR Analysis											
	Mean	Standard Deviation	Min Value	Max Value	Range	Aim	Upper SPC control limit	Lower SPC Control limit	Upper plant control limit	Lower plant control limit	Percentage of results out of plant tolerance
Auto	66.16	2.23	60.7	70.7	10.0	67	72.84	59.49		64.00	16.55%
Manual	66.3	2.195	59.4	71.8	12.4	67	72.89	59.72		64.00	12.05%
Difference	-0.14	0.035	1.3	-1.1	-2.4	0	-0.05	-0.23			4.50%

Figure 59 – Statistical table comparison of CSR results sampled manuallly and with the autosampler

The analysis results for CSR show very little change after implementation of the autosampler after two months. The mean results are very close as are the standard deviations, with a slight increase in standard deviation after using the autosampler to sample coke. The number of results out of specification has also slightly increased.

All results are within the SPC limits and so the process seems to be in control. There was one result out of control previously.

This suggests that despite fears over the sampling regime that the original manual method of sampling had little effect on CSR. Despite efforts from the coke ovens in improving heating stability and lowering Fe contamination by managing the stock yards, there has been little effect on the CSR results. There may be a need to revisit root cause analysis on the CSR variation in recent months.

Sample frequency could also be a consideration in the lack of change in these results. Currently, the coke is only sampled once every four hours and as already explained this represents only 8.3% of the ovens pushed daily. This can lead to overrepresentation of good and poor-quality coke in the sample and is not a true reflection of the quality of the coke being produced. To get a fully representative sample of the coke manufactured during a 24 hour period then a sample from every oven should be taken. This was the expectation when the project was started but full automation has not yet been achieved and the autosampler is being used manually. A fully representative sample would not guarantee the reduction in CSR standard deviation that the blast furnaces desire but there could at least be confidence in the results as the sampling and testing methods would then be following ISO standard recommendations.

CRI

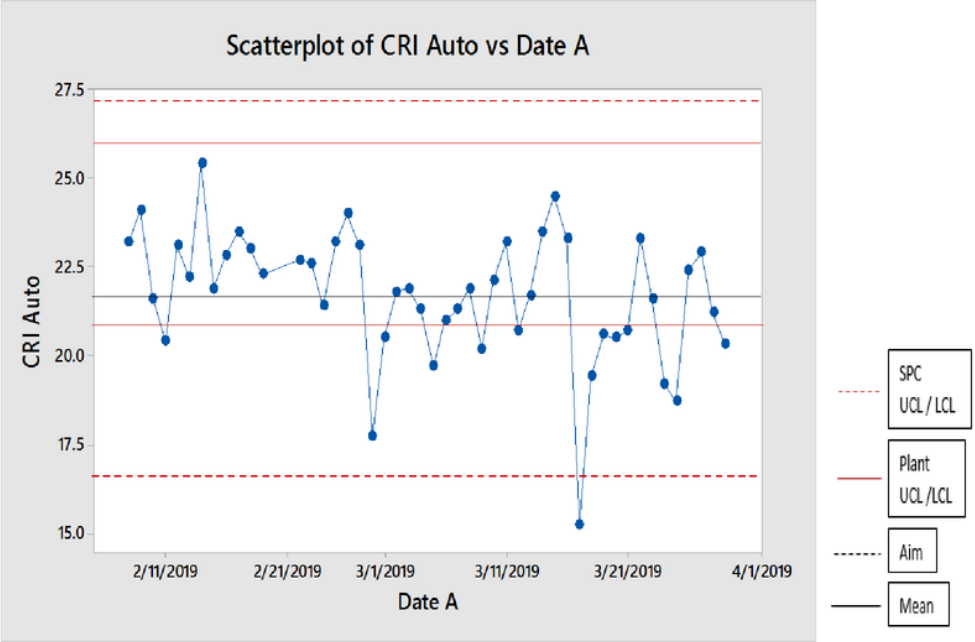


Figure 60 – SPC control chart of the CSR results from coke sampled from the autosampler

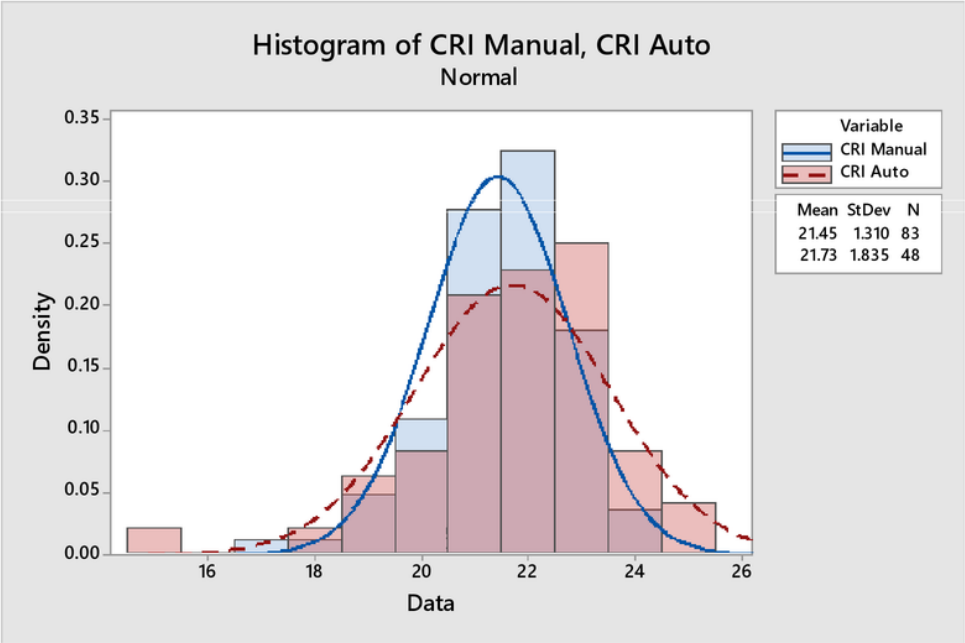


Figure 61 – Histogram and normal distribution comparison of CRI results sampled manuallly and with the autosampler

CRI Analysis											
	Mean	Standard Deviation	Min Value	Max Value	Range	Aim	Upper SPC control limit	Lower SPC Control limit	Upper plant control limit	Lower plant control limit	Percentage of results out of plant tolerance
Auto	21.73	1.84	15.2	25.4	10.2		27.24	16.68	≥ 21	≤ 26	35.63%
Manual	21.45	1.31	16.5	24.4	7.9		25.38	17.54	≥ 21	≤ 26	36.51%
Difference	0.28	0.53	-1.3	1.0	2.3		1.86	-0.86			-0.88%

Figure 62 – Statistical table comparison of CRI results sampled manuallly and with the autosampler

The CRI mean, as with the CSR mean, has not deviated much after changing sampling method, the amount of results outside the operational control limits also remains close. The standard deviation has increased by 0.53 from 1.31. This gain in standard deviation is unexpected and cannot be explained by the use of the autosampler and is more likely a result of process changes, more data should be collected to validate the result before investigating root causes.

There is one result outside of the SPC limits which was the same as the control chart for before the autosampler was introduced. The process seems in control most of the time. Although there are special causes present.

Moisture

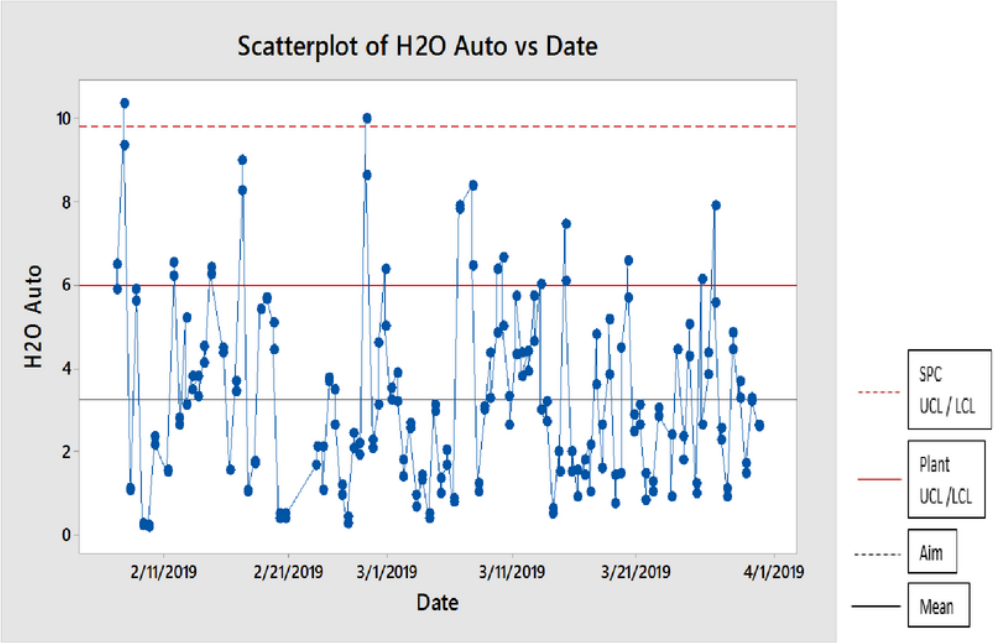


Figure 63 – SPC control chart of the moisture results from coke sampled from the autosampler

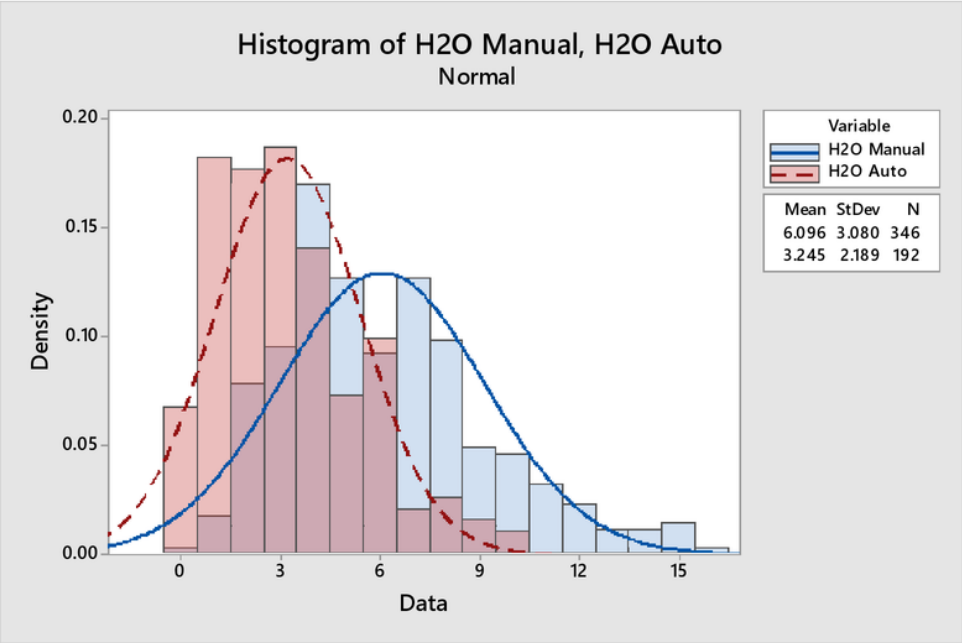


Figure 64 – Histogram and normal distribution comparison of moisture results sampled manuallly and with the autosampler

Moisture analysis											
	Mean	Standard Deviation	Min Value	Max Value	Range	Aim	Upper SPC control limit	Lower SPC Control limit	Upper plant control limit	Lower plant control limit	Percentage of results out of plant tolerance
Auto	3.25	2.19	0.18	10.34	10.16	< 6	9.812	0		6	11.90%
Manual	6.10	3.08	0.40	15.58	15.18	< 6	15.34	0		6	55.66%
Difference	-2.85	-0.89	-0.22	-5.24	-5.02		5.53				-43.76%

Figure 65 – Statistical table comparison of moisture results sampled manuallly and with the autosampler

There has been a significant drop in both the mean and standard variation since the change in sampling method, the mean has now fallen to well below the lower control limit and the number of results outside this limit has fallen by 43.76%. The change in mean cannot be attributed to the autosampler as a reduction in quench times was implemented at the start of the year (Dickinson, 2019). It could be said that the reduction in standard deviation may be attributed to the autosampler because human bias has been eliminated and it was suspected that samples were not being taken when the coke was steaming due to poor visibility, possibly leading to an over representation of dry or wet coke, more likely wet coke as sensors detect hot coke and activate cooling sprinklers to prevent burning it the conveyor (Dickinson, 2018).

There are two results seen outside of the SPC control limits, as with the pre autosampler data, this could be caused by ovens under filled with coal, resulting in over quenched coke since the amount of water added during quenching is constant. Ovens are often under filled when an oven has heating stability issues (Dickinson, 2019).

ASH

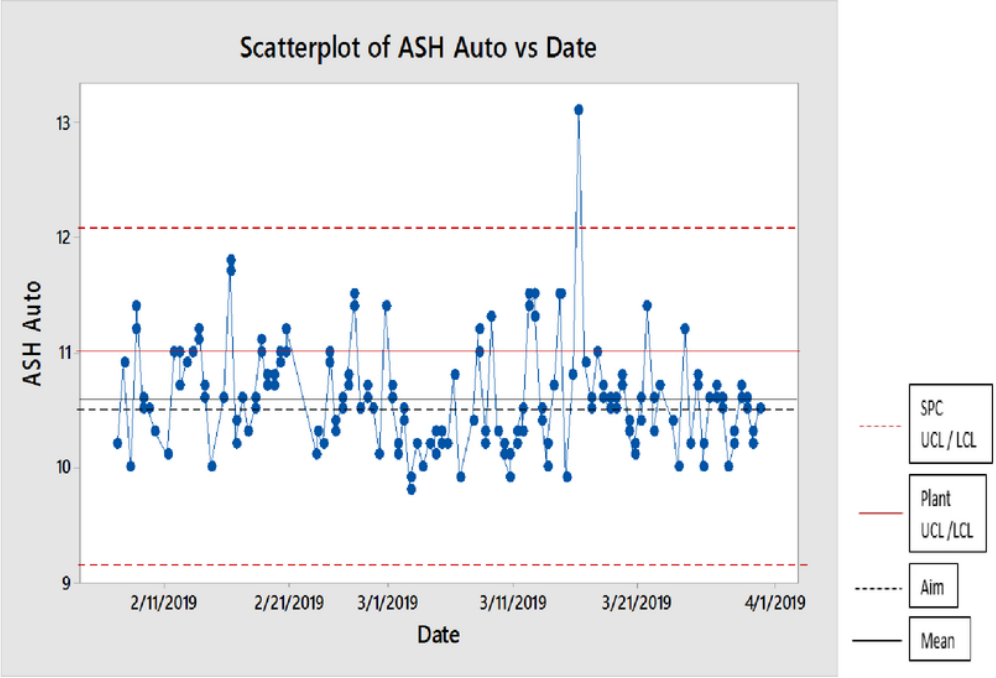


Figure 66 – SPC control chart of the ash results from coke sampled from the autosampler

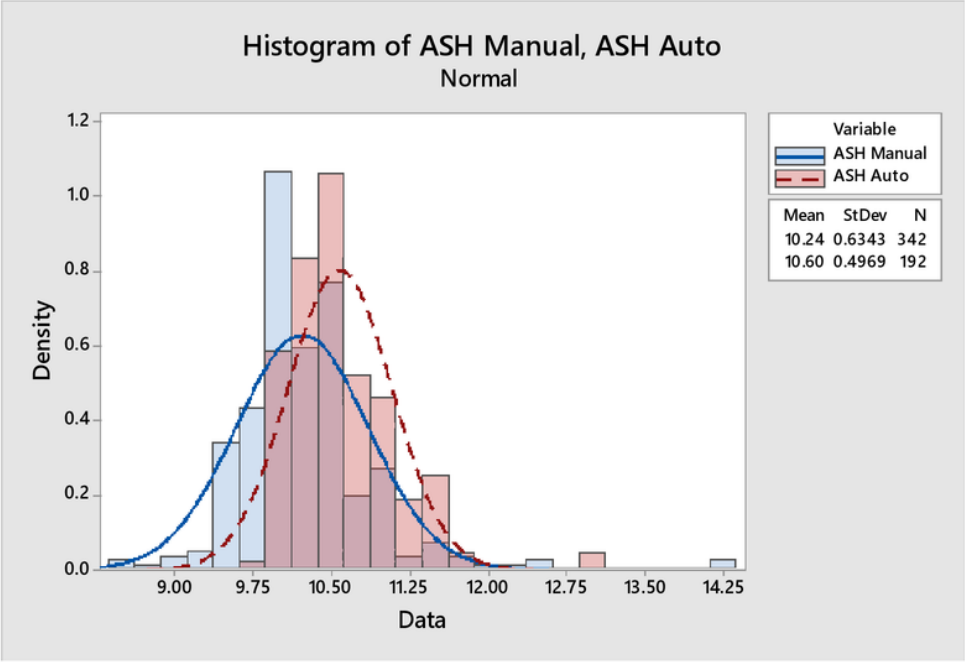


Figure 67 – Histogram and normal distribution comparison of ash results sampled manuallly and with the autosampler

Ash analysis											
	Mean	Standard Deviation	Min Value	Max Value	Range	Aim	Upper SPC control limit	Lower SPC Control limit	Upper plant control limit	Lower plant control limit	Percentage of results out of plant tolerance
Auto	10.60	0.50	9.81	13.10	3.29	≤ 10.5	12.09	9.11	11	9	20.90%
Manual	10.24	0.63	8.50	14.30	5.80	≤10.5	11.87	8.54	11	9	9.31%
Difference	0.36	-0.13	1.31	-1.2	2.51		0.22	0.57			11.59%

Figure 68 – Statistical table comparison of ash results sampled manuallly and with the autosampler

There has been a noticeable shift in mean for the ash results, although there has been no recent change in the composition of the coal blend there has been a number of small increases in ash noted in a number of the coals making up the blend, resulting in the increase seen in the mean (Dickinson, 2019). This could result in alterations made to the coal blend to correct this swing. This change has seen the number of results outside of the operation control limits rise 11.59%. There has however been a decrease in standard variation which may be a result of using the autosampler, a consequence of reducing the overrepresentation of under coked coal in samples.

There is only one result outside of the SPC tolerances, an improvement on the control chart for the manual method of sampling. This special cause is likely to be caused by Fe contamination in the coal (Díez, et al., 2002).

Mean size

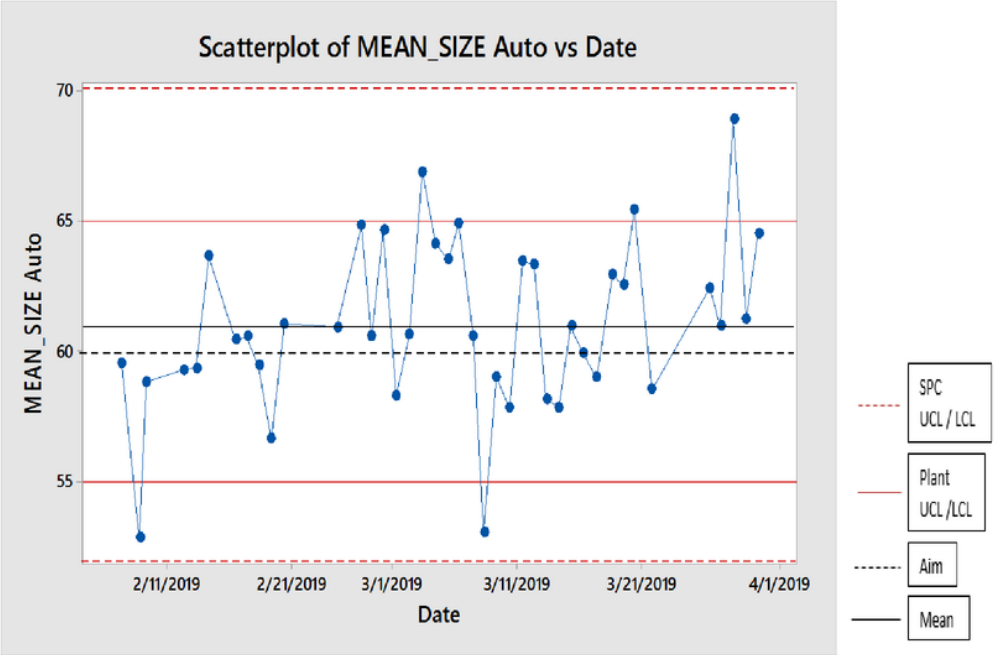


Figure 69 – SPC control chart of the mean size results from coke sampled from the autosampler

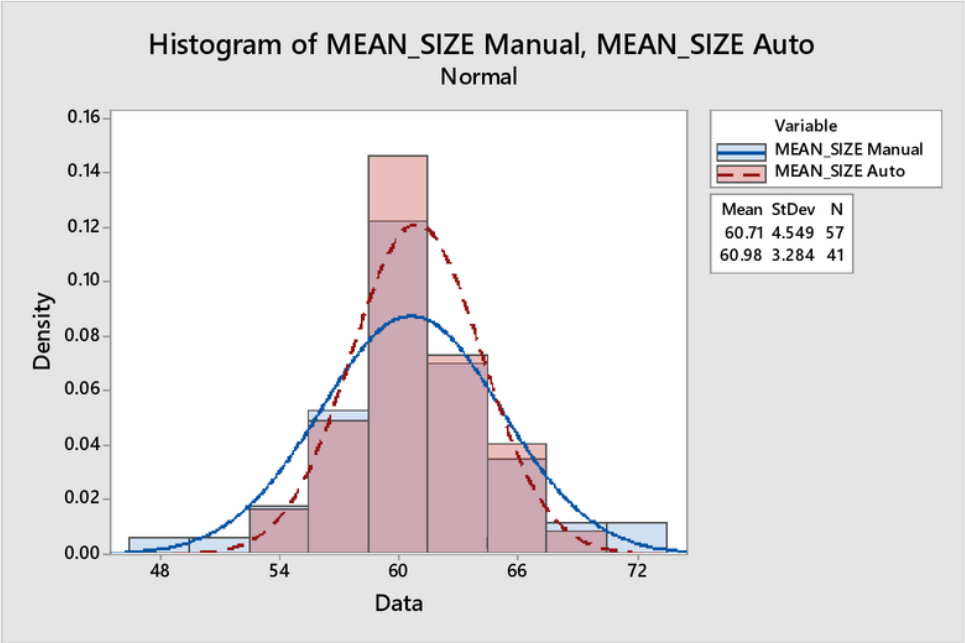


Figure 70 – Histogram and normal distribution comparison of mean size results sampled manuallly and with the autosampler

Mean size analysis											
	Mean	Standard Deviation	Min Value	Max Value	Range	Aim	Upper SPC control limit	Lower SPC Control limit	Upper plant control limit	Lower plant control limit	Percentage of results out of plant tolerance
Auto	60.982	3.28	52.79	68.85	16.06	60	70.83	51.13	65	55	14.48%
Manual	60.71	4.55	46.54	71.57	25.03	60	74.36	47.06	65	55	27.76%
Difference	0.27	-1.27	6.25	2.72	-8.97		-3.53	4.07			-13.28%

Figure 71 – Statistical table comparison of mean size results sampled manuallly and with the autosampler

Although the mean remains close after implementation of the autosampler there has been a considerable drop in the standard variation. The number of results outside the control limits has also dropped by 13.28%. One of the main contributors to coke size is soak time in the coke oven (Loison, et al., 1989), Morfa operates at an 18-hour soak time, this has remained consistent. It could be said that the main contributor to the drop in standard deviation is the use of the autosampler, as stockpile segregation is no longer an issue.

Micum M40

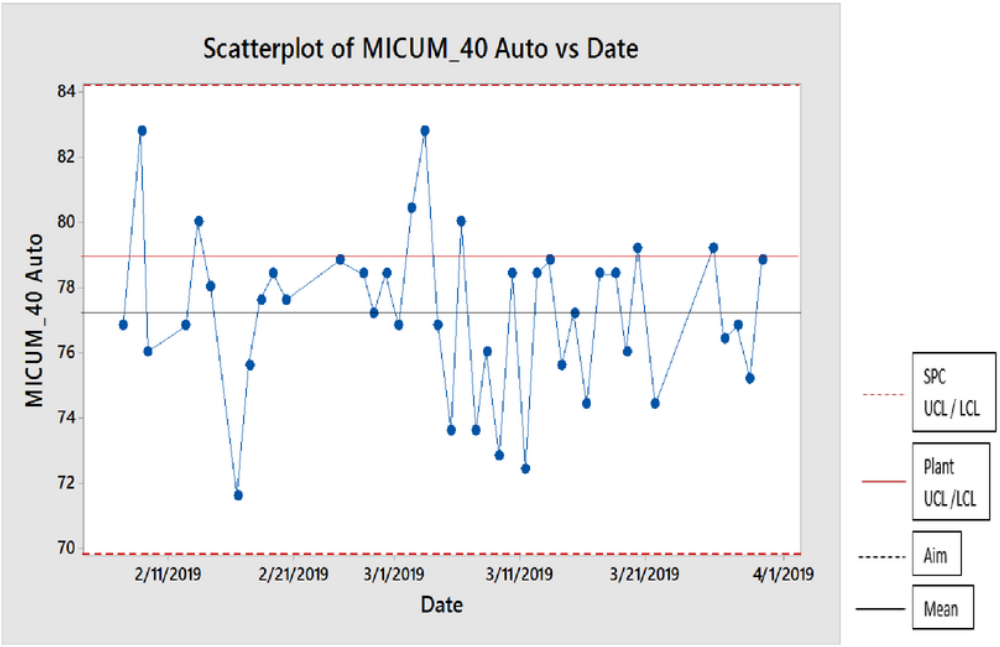


Figure 72 – SPC control chart of the M40 results from coke sampled from the autosampler

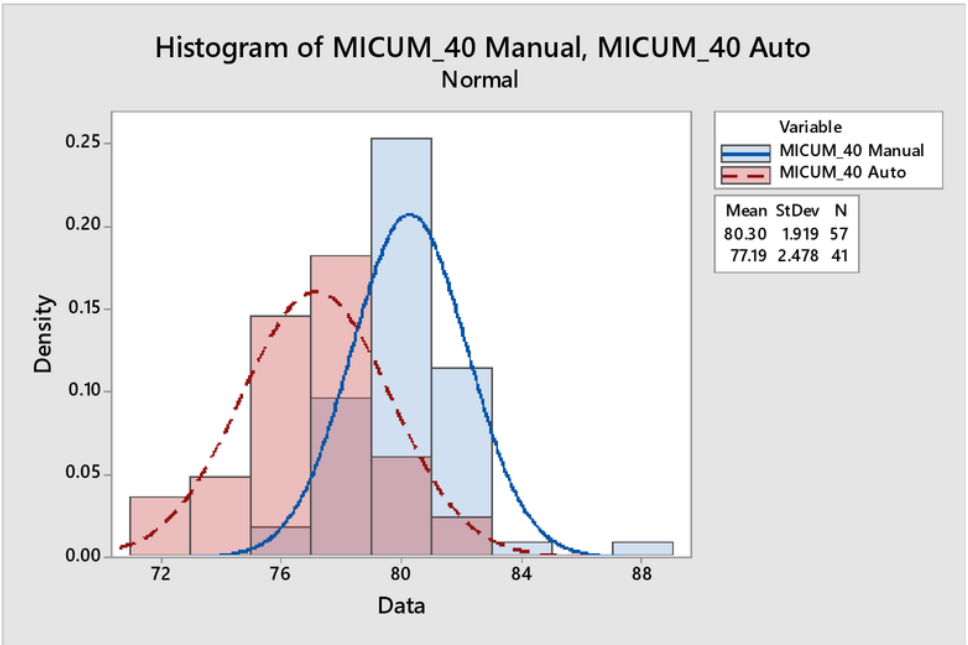


Figure 73 – Histogram and normal distribution comparison of M40 results sampled manuallly and with the autosampler

M40 analysis											
	Mean	Standard Deviation	Min Value	Max Value	Range	Aim	Upper SPC control limit	Lower SPC Control limit	Upper plant control limit	Lower plant control limit	Percentage of results out of plant tolerance
Auto	77.19	2.48	71.6	82.8	11.2	≥ 79	84.62	69.76		≥ 79	76.74%
Manual	80.3	1.92	76	88.8	12.8	≥ 79	86.06	74.54		≥ 79	24.62%
Difference	-3.11	0.56	-4.4	-6	-1.6		-1.44	-4.78			52.12%

Figure 74 – Statistical table comparison of M40 results sampled manuallly and with the autosampler

There has been a drop in the mean M40 result for the period between early January and late March, the most probable cause of this is are changes in the process, the micum comparison trial, run before implementation of the autosampler showed there was no significant change in M40 average after changing sampling methods. The coke ovens technical team originally attributed this drop in strength to the autosampler, until the trial carried out beforehand was presented to them. The mean has now dropped below the lower control limit of 79 and the number of results out of specification has risen by 52.12%. The coke ovens will need to investigate this shift in coke cold strength, as it will impact upon blast furnace stability.

This move in mean may have contributed to the increase in standard deviation, to confirm more data would need to be collected once the process has been stabilised and the mean returned to historic levels. It would also be worth re visit the method comparison trial to collect more data and confirm that the autosampler is not having a negative effect on cold strength.

There are no results outside of the SPC range and the process can be deemed in control, a slight improvement on the data collected for the manual method of sampling.

Micum 10

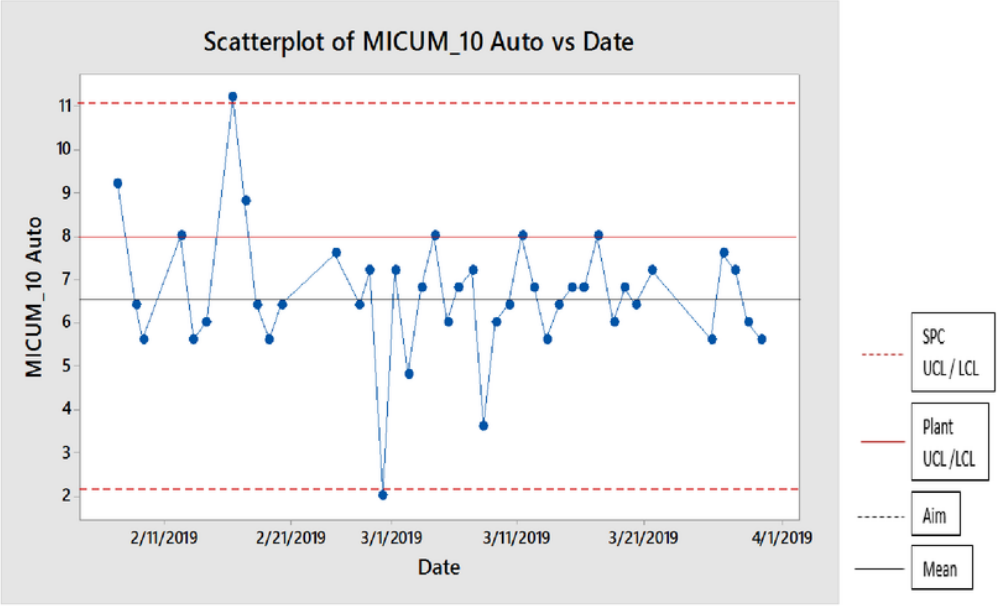


Figure 75 – SPC control chart of the M10 results from coke sampled from the autosampler

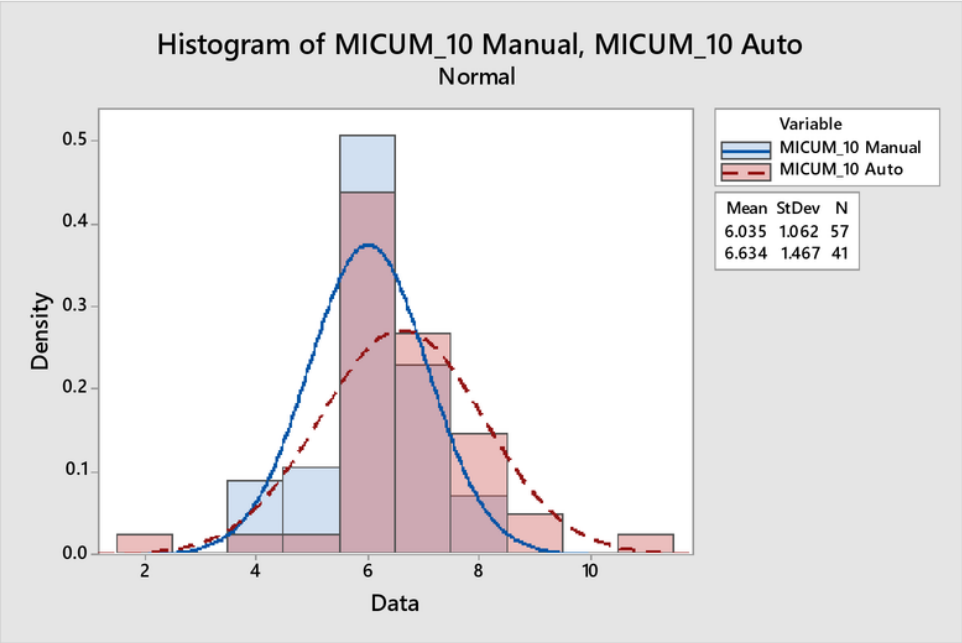


Figure 76 – Histogram and normal distribution comparison of M10 results sampled manuallly and with the autosampler

M10 analysis											
	Mean	Standard Deviation	Min Value	Max Value	Range	Aim	Upper SPC control limit	Lower SPC Control limit	Upper plant control limit	Lower plant control limit	Percentage of results out of plant tolerance
Auto	6.63	1.47	2.00	11.20	9.20	≤ 8	11.04	2.22	≤ 8		17.59%
Manual	6.04	1.06	3.60	8.40	4.80	≤ 8	9.22	2.86	≤ 8		4.09%
Difference	0.59	0.41	-1.6	2.8	4.4		1.82	-0.64			13.05

Figure 77 – Statistical table comparison of M10 results sampled manuallly and with the autosampler

The drop in quality seen in the M40 analysis is also shown in the M10 results. The mean has increased to 6.63 and the number of results outside of specification has increased by 13.05%. Although the mean is still well below the upper control limit. There has also been an increase in standard variation which may be explained by drop in coke quality and the fact that the sample frequency has remained the same.

There are two results outside of the SPC limits indicating the process is out of control, while was previously in control using the manual method of sampling.

Discussion and Conclusions

With concerns around the quality of metallurgical coke being produced at Morfa coke ovens, with CSR of particular interest. The CSR minimum specification had been raised to try increase the quality of the coke and reduce the likelihood of poor-quality coke reaching the blast furnaces and disrupt productivity. This increase in the CSR specification would amount to approximately £8 million per annum in increased coal blend costs. The method of sampling was identified as a possible source of variation and so an autosampler was purchased with the hope that this would eliminate any variation introduced through human error and bias. To try evaluate the impact of using the autosampler on coke quality, the results from the coke quality measures for hot strength and reactivity CSR/CSR, cold strength and abrasion M40/M10, ash, moisture and mean size were evaluated for periods before and after implementation of the autosampler. It was not possible to validate reduction in variation through use of the autosampler by comparing CSR results alone, as there were a number of ongoing initiatives with the common aim of CSR variation reduction. It was hoped that by demonstrating a reduction in all the quality measures, that it could be proved that the autosampler had a positive effect. This would in turn allow the reduction of the CSR specification limit back to historic levels.

It could be concluded from the data comparison, as there was no significant change in variation in CSR results after using the autosampler, that the manual method of sampling from the conveyor does not have any effect on the CSR result. It seems the sampling, preparation and testing of the coke is not a major factor in the poor quality of Morfa coke seen recently. The implementation of the autosampler, as yet, will not allow a change in the CSR lower specification back to historic levels.

There have been mixed results, some of the quality measures have seen drops in variation while some have seen an increase. The coke making process is complex with many variables, many of which can have an impact on coke quality. Some of the quality measure comparisons show that there may have been process shifts in the time between the two sets of data were collected, such as the coke quench time. The result of these process changes has meant that the mean has deviated in some of the quality measures, which is to be expected if there is a process shift, a change in standard deviation is a possible effect of a process change but it is probable the autosampler has had a positive effect in reducing the standard deviation in some of the quality measures. It is also possible that the autosampler has decreased variation in CSR but this is masked in the data by an increase in variation from another source.

The improvement in the mean size standard deviation is a confirmation that stockpile sampling is a poor method of sampling due to particle size segregation. ISO standards recommend using autosamplers for this reason along with eliminating random error and bias.

The increase in standard variation and drop in quality of the cold strength and abrasion index measures will be a concern for the coke ovens, the reasons for this change needs to be investigated, further testing will be required to reassess the introduction of the autosampler as a factor before coming to any accurate conclusions.

As previously discussed, the sample frequency remains an issue. To get a true representation of the coke quality over a 24 hour period then every oven pushed would need a sample taken from it. This would also allow an accurate measurement of the amount of variation inherent in the process. The current sample frequency of every four hours is adding unnecessary variation to coke quality analysis as there is an element of randomness to the ovens that are sampled. The coke quality from every oven will vary, the age of the battery also means that some of the ovens are in a poor condition which will negatively affect the coke quality produced from the oven.

The implementation of the autosampler aligns the sampling methods with ISO standards, which can only been seen as a positive, but to fully realise its potential to reduce standard variation, it will need to be used in a fully automatic mode, taking samples in 20 minute increments to try capture every oven pushed to ensure a fully representative sample for analysis.

Recommended Further Work

The full automation of the autosampler must be a priority to allow a fully representative sample. Once this is in place, then further comparison of the data would be required to assess the true impact of the autosampler on coke quality.

To enable full automation of the autosampler a receptacle for the storage of the extra coke resulting from the increased sample frequency. The amount of coke taken over 12 hours would be large, meaning the laboratory technicians would have difficulty transporting it back to the sample preparation building and also difficulty in mixing and splitting it due to the size of the sample and the large particle size of metallurgical coke. Therefore, equipment that could store, mix and split the coke at the autosampler would be advisable. Equipment that could mix, split and store the coke at the autosamplers location was sourced from a company in the Netherlands, see figure 78, a quote was obtained at 55,000 Euros. It is recommended that Tata purchase this equipment or a similar type of equipment that can carry out the necessary functions and install as soon as possible to enable representative sampling of Morfa coke.



Figure 78 - The big bag divider (Laarmann Group, 2018)

Another comparison of the manual method and the autosampler with samples taken at the same time is recommended, this time over a longer period to gather more data to enable a full assessment of any possible effects the autosampler has had on the M40/M10 results. The

downward trend in these results since implementation of the autosampler is a concern and any effects need to be fully understood.

Some experiments with the speed of the hammer that takes the autosampler is also recommended to see if any changes in its speed, will influence the M40/M10.

Consideration into the best location for the autosampler also needs to be taken. There are two options, the first is where the autosampler is located currently, at Morfa coke ovens. Another possible location is at the blast furnaces. Many steelmaking plants have their coke sample points at their blast furnaces. The argument for placing the sample point at the coke ovens is for better control of the coke making process, if the quality analysis shows poor quality coke has been made then the coke ovens can react and make changes and investigate the cause of the quality issues. The blast furnaces also will know in advance the quality of the coke on its way to the furnace and could decide not to use it if necessary. Coke sampled at the blast furnace has been fully stabilised on its journey there and the fines have also been screened out. The analysis of the coke sampled here gives a better understanding of the cokes' condition as it is charged into the furnace. Although if the quality of the coke is poor it is too late, as by the time the analysis comes back the coke will have been charged into the furnace (Sharma, et al., 2013).

References

Alakangas, E., 2015. *Quality guidelines of wood fuels in Finland,* Finland: Technical Research Centre of Finland VTT Ltd.

Álvarez, R. A. et al., 2007. An Approach to Blast Furnace Coke Quality Prediction. *Fuel,* 86(14), pp. 2159 - 2166.

Baradaran, V. & Dashtbani, H., 2014. A decision support system for monitoring traffic by statistical control charts. *International Journal of Industrial Engineering Computations,* 4(8), pp. 1661 - 1670.

BBC, 2016. *Tata Steel confirms 1,050 job cuts.* [Online]
Available at: www.bbc.co.uk/news/uk-wales-politics-35337896
[Accessed 17th November 2018].

British Standards Institute, 1992. *BS1016-108.2: 1992 Methods for Analysis and testing of coal and coke — Part108: Tests special to coke — Section 108.2 Determination of Micum and Irsid indices.* London: BSI.

British standards institute, 2009. *BS ISO 18283:2006 Hard Coal and Coke - Manual Sampling.* London: BSI Publications.

British Standards Institute, 2018. *BS ISO 18894:2018 Coke — Determination of coke reactivity index (CRI) and coke strength after reaction (CSR).* London: BSI.

British Standars Institute, 2000. *BS 1016-102:2000 ISO 579:1999 Methods for analysis and testing of coal and coke - Part 102: Determination of total moisture of coke.* London: BSI.

Danieli Corus BV, 2009. *Blast Furnace Operator Course developed for the use of Corus Port Talbot by Danieli Corus BV.* Port Talbot: Corus.

Dickinson, P., 2018. *Presenting at the Iron and Steel Technical Review.* Port Talbot, Tatasteel.

Dickinson, P., 2019. *Technical Manager Morfa Coke Ovens* [Interview] (29th March 2019).

Díez, M. A., Alvarez, R. & Barriocanal, C., 2002. Coal for Metallurgical Coke Production: Predictions of Coke Quality and Future Requirements for Cokemaking. *International Journal of Coal Geology,* 50(1 - 4), pp. 389 - 412.

Geerdes, M., Toxopeus, H. & Vliet, C. v. d., 2009. *Modern Blast Furnace Ironmaking an Introduction.* 2nd ed. Amsterdam: IOS press.

Haapakangas, J., 2016. Coke properties in Simulated Blast Furnace Conditions - Investigation on Hot Strength, Chemical Reactivity and Reaction Mechanism. *Academic dissertation to be presented, with the assent of the Doctoral Training Committee of Technology and Natural Sciences of the University of Oulu.*

Han, B., 1993. Studies of Metallurgical Coke Behaviour and Properties Under Conditions Relevant to the Bosh and Deadman Regions of a Blast Furnace. *A Doctoral Thesis.*

Hilding, T., 2005. *Evolution of Coke Properties while Descending through a Blast Furnace.* s.l.:Luleå University of Technology Department of Chemical Engineering and Geosciences Division of Process Metallurgy .

ISO, 2001. *Statistical aspects of sampling from bulk materials — Part 2: Sampling of particulate materials.* [Online]
Available at: https://www.iso.org/obp/ui/#iso:std:iso:11648:-2:ed-1:v1:en
[Accessed 27th October 2018].

James, D., 2016. *Welsh Steel in Crisis.* [Online]
Available at: www.walesonline.co.uk/news/politics/port-talbot-steelworks-up-sale-11111029
[Accessed 17th November 2018].

Jin, G., 2018. *Normal Distribution.* [Online]
Available at: http://my.ilstu.edu/~gjin/hsc204-hed/Module-5-Summary-Measure-2/Module-5-Summary-Measure-28.html
[Accessed 17th March 2019].

Laarmann Group, 2018. *Big Bag Divider.* The Netherlands: s.n.

Li, K. et al., 2014. The Evolution of Structural Order, Microstructure and Mineral Matter of Metallurgical Coke in a Blast Furnace: A Review. *Fuel,* 133(1), pp. 194 - 215.

Loison, R., Foch, P. & Boyer, A., 1989. *Coke Quality and Production.* Second ed. London: Butterworths.

Lundgren, M., 2010. Blast Furnace Coke Properties and the Influence on Off-gas Dust. *Licentiate Thesis.*

Lyman, G. et al., 2010. Bias testing of cross-belt samplers. *The Journal of The Southern African Institute of Mining and Metallurgy,* Volume 110, pp. 289 - 298.

Membrey, W., 2013. *The Coal Handbook: Towards Cleaner Production.* Australia: Woodhead publishing.

METaL, 2016. *Morfa Coke Oven Battery.* Swansea University: Materials Education Training and Learning.

Mortenson, P. P., 2015. *Representative Sampling 30 min crash course.* [Online]
Available at: https://www.teknologisk.dk/_/media/62672_Peter%20Mortensen%20-%20Representative%20Sampling%20-%2030%20min%20crash%20course.pdf
[Accessed 5th Febuary 2019].

Nag, D., Haldar, S. K., Choudhary, P. K. & Banerjee, P. K., 2009. Prediction of Coke CSR from Ash Chemistry of Coal Blend. *International Journal of Coal Preparation and Utilization,* 29(5), pp. 243 - 250.

Nenuwa, O. B. & Adetiloye, A., 2016. An Assessment of Laboratory Sample Splitting Techniques. *International Journal of Engineering and Technology,* 6(10), pp. 358 - 364.

Nohl, J. & Domnick, B., 2000. *Stockpile Segragation,* Minnesota: National Institute of Industrial Technology.

Robinson, G. K., Sinnott, M. D. & Cleary, P. W., 2010. Summary of results of ACARP project on cross-belt cutters. *The Journal of The Southern African Institute of Mining and Metallurgy,* Volume 110, pp. 331 - 338.

Russel, R. S. & Taylor III, B. W., 2011. *Operations Management Creating Value Along The Supply Chain.* Seventh ed. New Jersey: John Wiley & Sons, INC.

Sharma, R., Dharmendrab, K., Tiwaria, H. P. & Banerjee, P. K., 2013. Study on Coke Size Degradation from Coke Plant Wharf to Blast Furnaces Stock House. *Coke and Chemistry,* 56(11), pp. 412 - 418.

Slack, N., Brandon-Jones, A. & Johnston, R., 2013. *Operation Management.* Seventh ed. Endinborough Gate: Pearson Education Limited.

Statistics How To, 2019. *Statistics How To.* [Online]
Available at: www.statisticshowto.datasciencecentral.com/probability-and-statistics/standard-deviation/
[Accessed 17th March 2019].

Steinhaus, R. C. & Minnitt, R. C., 2014. Mechanical sampling - a manufacturer's perspective. *Journal of the Southern African Institute of Mining and Metallurgy,* 114(1).

Sympatec GbmH, 2018. *Sampling and Sample Splitting.* [Online]
Available at: www.sympatec.com/en/particle-measurement/glossary/sampling/
[Accessed 7th Febuary 2019].

Tatasteel, 2018. *Cost of Liquid steel - Wave 13 - Steering meeting#1.* Port Talbot: Tatasteel.

Tatasteel, 2019. *Port Talbot Coke Making - Blast Furnace Service Level Agrement.* Port Talbot Steelworks: Document: JDEY-5ADK2N.

Towsey, P. S., Cameron, I. & Gordon, Y., 2010. Comparison of Byproduct and Heat-Recovery Cokemaking Technologies. *The Iron & Steel Technology Conference and Exposition,* pp. 42 - 50.

United States Environmental Protection Agency, 1995. *AP-42: Compilation of Air Emissions Factors.* [Online]
Available at: www3.epa.gov/ttnchie1/ap42/ch12/final/c12s02_may08.pdf
[Accessed 10th Febuary 2019].

Waters, J., 2018. *Personal Figures and Photographs.* Port Talbot: Tatasteel.

Wuxi YORK Corporation, 2018. *Cross Belt Sampler Manual.* Shanghai: Wuxi YORK Corporation.

Abstract

Raw data for CSR and CRI before autosampler

Date	CRI	CSR	Date	CRI	CSR	Date	CRI	CSR	Date	CRI	CSR
02/10/2018	20.7	66.80	23/10/2018	21.2	66.90	18/11/2018	22.5	66.70	10/12/2018	23.0	63.21
03/10/2018	21.6	66.10	25/10/2018	22.1	63.80	19/11/2018	19.9	68.40	11/12/2018	22.2	65.70
04/10/2018	21.4	64.00	26/10/2018	20.5	67.90	20/11/2018	21.8	67.20	12/12/2018	20.3	66.00
05/10/2018	19.9	69.10	27/10/2018	21.1	67.60	21/11/2018	21.3	66.81	13/12/2018	21.2	66.70
06/10/2018	21.6	67.50	28/10/2018	21.6	65.30	22/11/2018	19.3	69.86	14/12/2018	21.2	66.00
07/10/2018	21.1	68.20	29/10/2018	20.5	66.00	23/11/2018	22.1	67.24	15/12/2018	23.0	66.40
08/10/2018	20.0	65.80	30/10/2018	21.5	66.00	24/11/2018	19.9	69.28	16/12/2018	23.1	65.00
09/10/2018	16.5	71.40	31/10/2018	20.7	68.70	25/11/2018	21.6	65.40	17/12/2018	23.5	65.60
10/10/2018	19.4	65.80	02/11/2018	21.1	67.30	26/11/2018	21.5	65.50	18/12/2018	21.5	62.10
11/10/2018	20.4	67.50	03/11/2018	21.7	64.70	27/11/2018	21.7	66.10	19/12/2018	22.1	64.30
12/10/2018	19.4	69.50	04/11/2018	23.1	62.50	28/11/2018	22.9	62.80	20/12/2018	24.4	59.40
13/10/2018	20.9	67.60	05/11/2018	22.0	64.40	29/11/2018	20.6	69.80	21/12/2018	23.2	64.10
14/10/2018	19.9	71.80	07/11/2018	22.4	67.70	30/11/2018	23.0	69.40	22/12/2018	23.1	61.90
15/10/2018	21.3	69.30	10/11/2018	18.2	66.15	01/12/2018	22.6	66.90	23/12/2018	21.8	65.40
17/10/2018	20.1	68.50	11/11/2018	21.0	65.90	02/12/2018	20.6	66.50	24/12/2018	21.0	67.75
18/10/2018	20.9	68.30	12/11/2018	23.1	63.51	04/12/2018	20.1	68.10	25/12/2018	23.4	65.49
19/10/2018	20.6	68.10	13/11/2018	22.4	63.73	05/12/2018	18.8	69.60	26/12/2018	23.2	66.73
20/10/2018	22.5	66.10	14/11/2018	21.7	66.18	06/12/2018	21.1	66.20	28/12/2018	22.0	61.30
21/10/2018	22.0	65.40	15/11/2018	22.3	65.87	07/12/2018	20.5	64.10	29/12/2018	22.2	65.40
22/10/2018	21.5	67.40	16/11/2018	21.8	64.69	08/12/2018	21.8	65.20	30/12/2018	21.7	68.00
			17/11/2018	21.4	67.60	09/12/2018	23.2	65.90	31/12/2018	23.6	65.70

Raw data for moisture and ash before autosampler

Date	H2O	ASH	Date	H2O	ASH	Date	H2O	ASH	Date	H2O	ASH	Date	H2O	ASH	Date	H2O	ASH
01/10/2018	4.5	9.6	06/10/2018	9.5	10.1	12/10/2018	2.0	10.0	18/10/2018	2.1	10.4	24/10/2018	2.7	10.9	29/10/2018	3.6	9.8
01/10/2018	4.2	9.6	06/10/2018	7.6	10.0	12/10/2018	1.0	9.8	19/10/2018	4.0	10.9	24/10/2018	3.3	10.8	29/10/2018	2.0	9.9
01/10/2018	9.6	9.7	06/10/2018	8.6	10.1	12/10/2018	10.7	10.0	19/10/2018	4.8	11.0	24/10/2018	9.6	10.4	29/10/2018	1.9	10.1
01/10/2018	9.8	9.9	07/10/2018	2.5	9.9	12/10/2018	15.3	9.8	19/10/2018	6.7	8.6	24/10/2018	11.3	10.4	30/10/2018	8.7	10.0
02/10/2018	4.5	10.1	07/10/2018	2.2	10.0	13/10/2018	4.2	10.0	19/10/2018	6.0	8.5	25/10/2018	7.6	11.0	30/10/2018	2.7	10.0
02/10/2018	4.8	10.1	07/10/2018	8.5	10.2	13/10/2018	4.3	10.1	20/10/2018	5.5	10.6	25/10/2018	5.2	10.9	31/10/2018	3.8	11.6
02/10/2018	2.7	10.9	07/10/2018	7.9	10.1	13/10/2018	2.5	10.8	20/10/2018	5.2	10.7	25/10/2018	4.4	11.8	31/10/2018	2.4	11.4
02/10/2018	4.4	10.8	08/10/2018	2.9	10.5	13/10/2018	2.3	10.6	20/10/2018	2.5	11.0	25/10/2018	5.0	11.3	31/10/2018	6.0	10.9
03/10/2018	5.5	10.0	08/10/2018	2.7	10.5	14/10/2018	5.7	11.4	20/10/2018	2.7	11.0	26/10/2018	11.2	10.8	31/10/2018	5.1	10.6
03/10/2018	4.7	10.2	08/10/2018	11.2	10.7	14/10/2018	5.1	11.3	21/10/2018	3.9	10.2	26/10/2018	8.2	10.8	01/11/2018	2.2	10.6
03/10/2018	3.7	10.3	08/10/2018	11.3	10.8	14/10/2018	4.1	10.6	21/10/2018	3.7	10.3	26/10/2018	10.8	10.3	01/11/2018	1.8	10.6
03/10/2018	4.3	10.2	09/10/2018	7.7	10.0	14/10/2018	6.7	10.7	21/10/2018	2.2	10.2	26/10/2018	7.8	10.2	01/11/2018	3.6	11.1
04/10/2018	6.8	9.4	09/10/2018	5.8	9.8	15/10/2018	3.5	10.4	21/10/2018	2.1	10.2	27/10/2018	6.4	11.0	01/11/2018	3.6	11.4
04/10/2018	6.8	9.4	09/10/2018	6.3	9.8	15/10/2018	5.1	10.6	22/10/2018	5.8	10.1	27/10/2018	6.6	10.9	02/11/2018	1.6	10.4
04/10/2018	8.4	10.2	09/10/2018	6.7	10.0	17/10/2018	5.2	10.9	22/10/2018	5.7	10.1	27/10/2018	7.1	10.5	02/11/2018	2.5	10.5
04/10/2018	7.2	9.9	10/10/2018	5.7	9.9	17/10/2018	4.3	10.6	22/10/2018	7.0	10.7	27/10/2018	5.6	10.4	02/11/2018	3.6	10.5
05/10/2018	3.4	9.9	10/10/2018	6.0	10.0	17/10/2018	4.1	10.4	22/10/2018	6.7	10.6	28/10/2018	11.8	11.1	02/11/2018	3.4	10.6
05/10/2018	3.2	9.9	10/10/2018	4.9	10.1	17/10/2018	5.1	10.6	23/10/2018	4.5	10.6	28/10/2018	10.9	11.1	03/11/2018	4.7	10.5
05/10/2018	4.9	11.8	10/10/2018	5.1	10.2	18/10/2018	14.2	10.7	23/10/2018	4.2	10.6	28/10/2018	7.9	10.6	03/11/2018	6.3	10.8
05/10/2018	4.2	11.7	11/10/2018	1.5	10.0	18/10/2018	13.7	10.8	23/10/2018	6.9	9.9	28/10/2018	7.9	10.4	03/11/2018	1.8	11.0
06/10/2018	10.0	10.1	11/10/2018	1.6	10.2	18/10/2018	2.3	10.5	23/10/2018	5.3	9.8	29/10/2018	3.3	9.7	03/11/2018	1.0	10.9

Date	H2O	ASH	Date	H2O	ASH	Date	H2O	ASH	Date	H2O	ASH	Date	H2O	ASH	Date	H2O	ASH
04/11/2018	2.6	10.1	11/11/2018	5.3	9.9	16/11/2018	8.7	9.6	22/11/2018	5.2	10.5	27/11/2018	4.1	10.3	03/12/2018	4.0	10.0
04/11/2018	1.8	10.0	11/11/2018	7.1	10.0	16/11/2018	6.6	9.6	22/11/2018	5.9	10.5	27/11/2018	4.6	10.6	04/12/2018	6.9	9.8
04/11/2018	2.6	10.0	11/11/2018	6.6	10.0	17/11/2018	10.0	9.9	22/11/2018	7.1	10.4	28/11/2018	3.6	9.9	04/12/2018	9.2	10.1
04/11/2018	2.5	9.9	12/11/2018	9.4	10.2	17/11/2018	9.2	9.9	23/11/2018	6.6	10.5	28/11/2018	3.2	9.9	04/12/2018	7.2	10.1
05/11/2018	2.9	9.7	12/11/2018	8.3	10.2	17/11/2018	3.6	10.4	23/11/2018	6.7	10.5	28/11/2018	2.4	9.5	04/12/2018	7.3	10.2
05/11/2018	2.9	9.8	12/11/2018	7.1	9.3	17/11/2018	2.8	10.5	23/11/2018	7.5	10.3	28/11/2018	3.9	9.4	05/12/2018	5.9	14.3
05/11/2018	5.0	8.9	12/11/2018	7.9	9.3	18/11/2018	5.8	10.9	23/11/2018	6.9	10.4	29/11/2018	2.5	10.9	05/12/2018	5.6	14.2
05/11/2018	4.9	8.7	13/11/2018	9.9	9.5	18/11/2018	9.1	9.9	24/11/2018	12.9	9.9	29/11/2018	3.0	11.0	05/12/2018	8.3	10.6
06/11/2018	2.4	10.1	13/11/2018	12.2	9.6	18/11/2018	14.2	9.6	24/11/2018	14.9	9.9	30/11/2018	6.2	10.0	05/12/2018	9.6	10.5
06/11/2018	3.3	10.1	13/11/2018	7.7	9.7	18/11/2018	12.4	9.8	24/11/2018	7.2	9.9	30/11/2018	6.8	10.2	06/12/2018	4.9	9.1
08/11/2018	5.6	10.6	13/11/2018	7.5	9.8	19/11/2018	8.6	10.5	24/11/2018	9.6	9.9	30/11/2018	4.2	9.7	06/12/2018	5.4	9.3
08/11/2018	4.6	10.6	14/11/2018	4.4	10.0	19/11/2018	6.6	10.6	25/11/2018	6.8	9.9	30/11/2018	4.6	9.7	06/12/2018	5.8	10.2
08/11/2018	2.8	10.2	14/11/2018	4.4	10.1	20/11/2018	3.7	10.2	25/11/2018	5.1	10.0	01/12/2018	5.3	10.1	06/12/2018	5.6	10.2
08/11/2018	1.8	10.3	14/11/2018	5.5	9.9	20/11/2018	4.3	10.1	25/11/2018	7.3	10.0	01/12/2018	5.8	9.9	07/12/2018	5.3	10.4
09/11/2018	2.4	10.6	14/11/2018	8.7	10.0	20/11/2018	10.4	10.2	25/11/2018	8.3	10.1	01/12/2018	5.7	9.2	07/12/2018	6.1	10.4
09/11/2018	2.2	10.6	15/11/2018	6.6	9.7	20/11/2018	8.4	10.5	26/11/2018	11.2	9.7	01/12/2018	4.5	9.1	07/12/2018	10.5	9.9
10/11/2018	11.8	9.5	15/11/2018	7.0	9.9	21/11/2018	3.7	10.1	26/11/2018	13.0	9.7	02/12/2018	7.9	10.0	07/12/2018	10.0	9.9
10/11/2018	12.9	9.4	15/11/2018	9.2	9.6	21/11/2018	4.3	10.2	26/11/2018	9.6	12.0	02/12/2018	8.3	10.1	08/12/2018	4.1	9.7
10/11/2018	3.3	9.7	15/11/2018	7.2	9.6	21/11/2018	7.1	10.4	26/11/2018	9.1	12.2	03/12/2018	4.6	10.2	08/12/2018	4.4	9.5
10/11/2018	2.7	9.8	16/11/2018	11.3	11.0	21/11/2018	7.0	10.4	27/11/2018	4.2	10.2	03/12/2018	4.0	10.3	08/12/2018	5.2	9.6
11/11/2018	4.6	9.8	16/11/2018	11.7	10.9	22/11/2018	5.3	10.6	27/11/2018	4.2	10.1	03/12/2018	4.6	10.0	08/12/2018	5.4	9.4

Date	H2O	ASH	Date	H2O	ASH	Date	H2O	ASH	Date	H2O	ASH	Date	H2O	ASH
09/12/2018	7.9	9.6	14/12/2018	2.4	10.4	19/12/2018	7.2	10.3	25/12/2018	1.7	10.3	30/12/2018	3.3	10.0
09/12/2018	7.8	9.4	14/12/2018	7.7	10.0	19/12/2018	6.6	10.3	25/12/2018	3.9	10.4	30/12/2018	2.9	10.4
09/12/2018	0.6	9.9	14/12/2018	7.7	10.2	20/12/2018	7.7	10.2	25/12/2018	4.3	10.4	31/12/2018	1.1	11.1
09/12/2018	0.4	10.2	15/12/2018	8.8	9.9	20/12/2018	6.9	10.2	26/12/2018	4.4	10.5	31/12/2018	1.3	11.3
10/12/2018	5.4	9.5	15/12/2018	9.2	10.1	21/12/2018	6.7	12.5	26/12/2018	4.1	10.4	31/12/2018	5.8	9.7
10/12/2018	4.4	9.4	15/12/2018	15.6	9.7	21/12/2018	6.3	12.5	26/12/2018	12.1	10.7	31/12/2018	7.2	9.7
10/12/2018	3.1	10.3	15/12/2018	15.2	9.8	21/12/2018	9.9	10.1	26/12/2018	8.2	10.7			
10/12/2018	4.3	10.3	16/12/2018	6.4	10.5	21/12/2018	9.4	10.2	27/12/2018	12.1	10.4			
11/12/2018	4.5	9.5	16/12/2018	7.7	10.4	22/12/2018	4.3	10.7	27/12/2018	8.2	10.2			
11/12/2018	4.3	10.0	16/12/2018	4.2	9.4	22/12/2018	4.6	10.6	27/12/2018	9.4	9.9			
11/12/2018	7.7	9.7	16/12/2018	7.9	9.4	22/12/2018	10.2	10.3	27/12/2018	8.8	10.0			
11/12/2018	10.0	9.7	17/12/2018	6.4	9.9	22/12/2018	10.7	10.3	28/12/2018	4.8	10.2			
12/12/2018	7.2	10.0	17/12/2018	8.1	10.1	23/12/2018	7.0	10.2	28/12/2018	4.1	10.4			
12/12/2018	7.7	9.6	17/12/2018	7.3	9.6	23/12/2018	7.4	10.2	28/12/2018	6.3	9.9			
12/12/2018	3.2	9.7	17/12/2018	7.0	9.5	23/12/2018	3.6	10.3	28/12/2018	8.0	10.2			
12/12/2018	3.6	9.7	18/12/2018	11.4	9.7	23/12/2018	3.3	10.2	29/12/2018	4.4	10.6			
13/12/2018	3.5	9.9	18/12/2018	11.5	9.5	24/12/2018	4.4	10.0	29/12/2018	5.7	10.7			
13/12/2018	3.7	9.7	18/12/2018	13.1	11.4	24/12/2018	4.3	9.7	29/12/2018	3.8	10.1			
13/12/2018	14.7	10.2	18/12/2018	14.2	11.4	24/12/2018	5.0	10.0	29/12/2018	3.9	9.8			
13/12/2018	14.7	10.4	19/12/2018	6.0	9.9	24/12/2018	7.5	10.3	30/12/2018	2.8	9.8			
14/12/2018	2.4	10.4	19/12/2018	4.6	10.0	25/12/2018	1.7	10.2	30/12/2018	1.9	9.8			

Raw data for mean size, M40 and M10 before autosampler

Date	MEAN_SIZE	MICUM_40	MICUM_10	Date	MEAN_SIZE	MICUM_40	MICUM_10	Date	MEAN_SIZE	MICUM_40	MICUM_10
10/20/2018	70.35	80.40	6.80	11/19/2018	60.52	79.20	4.80	12/14/2018	59.23	81.60	5.60
10/21/2018	70.93	81.60	6.80	11/20/2018	53.99	82.40	6.80	12/15/2018	59.44	80.40	5.20
10/22/2018	69.71	80.00	5.60	11/21/2018	53.79	80.00	6.40	12/16/2018	57.71	80.80	6.00
10/23/2018	57.47	79.20	3.60	11/22/2018	57.63	78.00	6.80	12/17/2018	60.12	76.00	5.60
10/24/2018	58.90	79.60	8.40	11/23/2018	71.57	82.00	7.20	12/18/2018	58.67	80.40	6.00
10/25/2018	65.06	80.00	5.60	11/24/2018	59.82	77.60	6.80	12/19/2018	57.96	80.80	3.60
10/29/2018	58.45	81.20	6.80	11/25/2018	60.51	84.00	4.00	12/22/2018	64.90	80.40	6.80
11/1/2018	58.45	79.20	8.40	11/26/2018	62.60	82.40	5.60	12/23/2018	62.01	80.00	6.00
11/2/2018	59.51	80.40	6.80	11/27/2018	59.70	81.60	6.00	12/24/2018	63.75	79.60	5.60
11/3/2018	62.09	80.80	5.60	11/28/2018	59.31	79.60	6.80	12/25/2018	62.59	78.80	6.40
11/4/2018	59.74	80.00	6.40	11/30/2018	64.14	78.40	7.20	12/26/2018	65.97	78.00	6.40
11/5/2018	61.63	82.80	7.60	12/1/2018	65.01	78.80	6.40	12/27/2018	64.84	79.20	6.00
11/6/2018	64.37	79.60	6.00	12/2/2018	64.98	78.80	7.20	12/28/2018	63.24	80.80	5.60
11/8/2018	56.38	81.60	5.20	12/3/2018	58.57	82.40	4.80	12/29/2018	56.26	80.40	6.00
11/10/2018	62.27	79.60	5.60	12/4/2018	50.89	78.40	7.20	12/31/2018	61.20	82.40	6.40
11/12/2018	59.72	76.80	7.60	12/5/2018	61.04	80.80	6.40				
11/14/2018	57.64	78.80	6.40	12/6/2018	63.02	80.00	5.60				
11/15/2018	61.42	78.80	5.60	12/7/2018	58.68	82.80	5.20				
11/16/2018	60.50	81.20	6.40	12/7/2018	52.96	88.80	4.00				
11/17/2018	46.54	80.00	4.80	12/8/2018	60.24	78.80	5.60				
11/18/2018	63.53	80.80	3.60	12/9/2018	59.05	80.40	6.40				

Raw data for CSR and CRI after autosampler

Date	CRI	CSR	Date	CRI	CSR	Date	CRI	CSR
08/02/2019	23.20	65.60	03/03/2019	21.90	65.80	24/03/2019	19.20	66.20
09/02/2019	24.10	66.10	04/03/2019	21.30	65.40	25/03/2019	18.70	68.40
10/02/2019	21.60	69.40	05/03/2019	19.70	68.60	26/03/2019	22.40	66.80
11/02/2019	20.40	65.30	06/03/2019	21.00	68.40	27/03/2019	22.90	64.30
12/02/2019	23.10	64.50	07/03/2019	21.30	67.60	28/03/2019	21.20	66.90
13/02/2019	22.20	64.50	08/03/2019	21.90	70.70	29/03/2019	20.30	68.30
14/02/2019	25.40	60.70	09/03/2019	20.20	65.90			
15/02/2019	21.90	65.90	10/03/2019	22.10	66.20			
16/02/2019	22.80	66.00	11/03/2019	23.20	63.80			
17/02/2019	23.50	66.40	12/03/2019	20.70	67.00			
18/02/2019	23.00	66.00	13/03/2019	21.70	70.40			
19/02/2019	22.30	65.60	14/03/2019	23.50	65.70			
22/02/2019	22.70	62.50	15/03/2019	24.50	62.10			
23/02/2019	22.60	61.80	16/03/2019	23.30	66.20			
24/02/2019	21.40	66.40	17/03/2019	15.20	62.40			
25/02/2019	23.20	63.40	18/03/2019	19.40	69.00			
26/02/2019	24.00	69.90	19/03/2019	20.60	69.00			
27/02/2019	23.10	65.90	20/03/2019	20.50	67.20			
28/02/2019	17.70	65.90	21/03/2019	20.70	64.70			
01/03/2019	20.50	69.00	22/03/2019	23.30	66.00			
02/03/2019	21.80	65.90	23/03/2019	21.60	66.10			

Raw data for moisture and ash after autosampler

Date	H2O	ASH	Date	H2O	ASH	Date	H2O	ASH	Date	H2O	ASH	Date	H2O	ASH	Date	H2O	ASH
07/02/2019	5.88	10.20	12/02/2019	3.10	10.90	18/02/2019	5.41	11.00	25/02/2019	0.40	10.80	03/03/2019	0.93	10.20	08/03/2019	3.06	10.20
07/02/2019	6.49	10.20	13/02/2019	3.79	11.00	18/02/2019	5.42	11.10	26/02/2019	2.44	11.50	03/03/2019	0.66	10.20	09/03/2019	4.34	11.30
07/02/2019	9.35	10.90	13/02/2019	3.48	11.00	19/02/2019	5.65	10.70	26/02/2019	2.08	11.40	03/03/2019	1.43	10.00	09/03/2019	3.28	11.30
07/02/2019	10.34	10.90	13/02/2019	3.78	11.10	19/02/2019	5.67	10.80	26/02/2019	2.20	10.50	03/03/2019	1.31	10.00	09/03/2019	4.85	10.30
08/02/2019	1.06	10.00	13/02/2019	3.29	11.20	19/02/2019	5.06	10.70	26/02/2019	1.89	10.50	04/03/2019	0.49	10.20	09/03/2019	6.36	10.30
08/02/2019	1.09	10.00	14/02/2019	4.50	10.60	19/02/2019	4.45	10.80	27/02/2019	9.97	10.60	04/03/2019	0.38	10.20	10/03/2019	6.64	10.20
08/02/2019	5.61	11.20	14/02/2019	4.11	10.70	20/02/2019	0.39	10.90	27/02/2019	8.63	10.70	04/03/2019	3.12	10.10	10/03/2019	4.99	10.10
08/02/2019	5.88	11.40	14/02/2019	6.40	10.00	20/02/2019	0.49	11.00	27/02/2019	2.05	10.50	04/03/2019	2.94	10.30	10/03/2019	3.29	9.90
09/02/2019	0.27	10.60	14/02/2019	6.26	10.00	20/02/2019	0.39	11.00	27/02/2019	2.26	10.50	05/03/2019	0.97	10.30	10/03/2019	2.62	10.10
09/02/2019	0.23	10.50	15/02/2019	4.35	10.60	20/02/2019	0.49	11.20	28/02/2019	3.10	10.10	05/03/2019	1.32	10.20	11/03/2019	4.31	10.20
09/02/2019	0.20	10.50	15/02/2019	4.48	10.60	23/02/2019	1.68	10.10	28/02/2019	4.60	10.10	05/03/2019	1.67	10.20	11/03/2019	5.73	10.30
09/02/2019	0.18	10.50	16/02/2019	1.53	11.70	23/02/2019	2.09	10.30	28/02/2019	6.36	11.40	05/03/2019	2.01	10.20	11/03/2019	4.34	10.50
10/02/2019	2.35	10.30	16/02/2019	1.53	11.80	23/02/2019	2.09	10.20	28/02/2019	4.98	11.40	06/03/2019	0.76	10.80	11/03/2019	3.81	10.30
10/02/2019	2.15	10.30	16/02/2019	3.44	10.40	23/02/2019	1.07	10.20	01/03/2019	3.21	10.70	06/03/2019	0.86	10.80	12/03/2019	4.40	11.50
11/02/2019	1.50	10.10	16/02/2019	3.68	10.20	24/02/2019	3.69	10.90	01/03/2019	3.53	10.60	06/03/2019	7.90	9.90	12/03/2019	3.90	11.40
11/02/2019	1.53	10.10	17/02/2019	8.24	10.60	24/02/2019	3.76	11.00	01/03/2019	3.88	10.20	06/03/2019	7.83	9.90	12/03/2019	5.73	11.50
11/02/2019	6.19	11.00	17/02/2019	8.99	10.60	24/02/2019	3.47	10.30	01/03/2019	3.18	10.10	07/03/2019	8.38	10.40	12/03/2019	4.62	11.30
11/02/2019	6.54	11.00	17/02/2019	1.00	10.30	24/02/2019	2.63	10.40	02/03/2019	1.79	10.50	07/03/2019	6.45	10.40	13/03/2019	6.00	10.40
12/02/2019	2.79	11.00	17/02/2019	1.04	10.30	25/02/2019	0.92	10.50	02/03/2019	1.36	10.40	08/03/2019	1.01	11.20	13/03/2019	3.00	10.50
12/02/2019	2.61	10.70	18/02/2019	1.76	10.50	25/02/2019	1.19	10.60	02/03/2019	2.65	9.80	08/03/2019	1.22	11.00	13/03/2019	2.71	10.00
12/02/2019	5.20	10.90	18/02/2019	1.69	10.60	25/02/2019	0.25	10.70	02/03/2019	2.54	9.90	08/03/2019	2.98	10.30	13/03/2019	3.18	10.20

Date	H2O	ASH	Date	H2O	ASH	Date	H2O	ASH	Date	H2O	ASH
14/03/2019	0.48	10.70	19/03/2019	1.42	10.60	25/03/2019	5.05	10.20	30/03/2019	3.19	10.30
14/03/2019	0.60	10.70	19/03/2019	1.48	10.70	25/03/2019	4.27	10.20	30/03/2019	2.62	10.50
14/03/2019	2.00	11.50	19/03/2019	4.47	10.80	25/03/2019	0.98	10.70	30/03/2019	2.58	10.50
14/03/2019	1.50	11.50	20/03/2019	5.70	10.40	25/03/2019	1.20	10.80			
15/03/2019	7.45	9.90	20/03/2019	6.56	10.30	26/03/2019	6.14	10.20			
15/03/2019	6.08	9.90	20/03/2019	2.46	10.10	26/03/2019	2.64	10.00			
15/03/2019	1.50	10.80	20/03/2019	2.88	10.20	26/03/2019	3.83	10.60			
15/03/2019	2.00	10.80	21/03/2019	3.09	10.60	26/03/2019	4.35	10.60			
16/03/2019	0.89	13.10	21/03/2019	2.62	10.40	27/03/2019	7.88	10.60			
16/03/2019	1.55	13.10	21/03/2019	0.83	11.40	27/03/2019	5.57	10.70			
16/03/2019	1.43	10.90	21/03/2019	1.44	11.40	27/03/2019	2.26	10.50			
16/03/2019	1.79	10.90	22/03/2019	1.27	10.60	27/03/2019	2.56	10.60			
17/03/2019	2.14	10.60	22/03/2019	1.00	10.30	28/03/2019	0.89	10.00			
17/03/2019	1.02	10.50	22/03/2019	3.01	10.70	28/03/2019	1.10	10.00			
17/03/2019	4.80	11.00	22/03/2019	2.83	10.70	28/03/2019	4.83	10.20			
17/03/2019	3.60	11.00	23/03/2019	2.37	10.40	28/03/2019	4.45	10.30			
18/03/2019	1.58	10.70	23/03/2019	0.91	10.40	29/03/2019	3.28	10.60			
18/03/2019	2.63	10.60	24/03/2019	4.45	10.00	29/03/2019	3.67	10.70			
18/03/2019	3.82	10.50	24/03/2019	4.43	10.00	29/03/2019	1.72	10.50			
18/03/2019	5.16	10.60	24/03/2019	2.36	11.20	29/03/2019	1.47	10.60			
19/03/2019	0.74	10.50	24/03/2019	1.79	11.20	30/03/2019	3.28	10.20			

Raw data for mean size, M40 and M10 after autosampler

Date	MEAN_SIZE	MICUM_40	MICUM_10	Date	MEAN_SIZE	MICUM_40	MICUM_10
07/02/2019	59.49	76.80	9.20	07/03/2019	60.56	73.60	7.20
08/02/2019	52.79	82.80	6.40	08/03/2019	52.98	76.00	3.60
09/02/2019	58.79	76.00	5.60	09/03/2019	58.99	72.80	6.00
12/02/2019	59.25	76.80	8.00	10/03/2019	57.79	78.40	6.40
13/02/2019	59.33	80.00	5.60	11/03/2019	63.46	72.40	8.00
14/02/2019	63.63	78.00	6.00	12/03/2019	63.29	78.40	6.80
16/02/2019	60.43	71.60	11.20	13/03/2019	58.13	78.80	5.60
17/02/2019	60.52	75.60	8.80	14/03/2019	57.80	75.60	6.40
18/02/2019	59.43	77.60	6.40	15/03/2019	60.94	77.20	6.80
19/02/2019	56.60	78.40	5.60	16/03/2019	59.86	74.40	6.80
20/02/2019	61.01	77.60	6.40	17/03/2019	58.94	78.40	8.00
24/02/2019	60.90	78.80	7.60	18/03/2019	62.93	78.40	6.00
26/02/2019	64.79	78.40	6.40	19/03/2019	62.49	76.00	6.80
27/02/2019	60.54	77.20	7.20	20/03/2019	65.38	79.20	6.40
28/02/2019	64.61	78.40	2.00	21/03/2019	58.49	74.40	7.20
01/03/2019	58.28	76.80	7.20	26/03/2019	62.39	79.20	5.60
02/03/2019	60.60	80.40	4.80	27/03/2019	60.91	76.40	7.60
03/03/2019	66.86	82.80	6.80	28/03/2019	68.85	76.80	7.20
04/03/2019	64.11	76.80	8.00	29/03/2019	61.22	75.20	6.00
05/03/2019	63.50	73.60	6.00	30/03/2019	64.47	78.80	5.60
06/03/2019	64.90	80.00	6.80				