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Verification of ideal semi-logarithmic, lognormal or fractal crystal size distributions from 2D datasets

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Abstract

Crystal size distributions can follow any function, provided the total crystal content is less than 100%. However, many theories suggest that they approximate a number of ideal distribution models. The most widely adopted model for igneous rocks produces a straight line on a 'classic' CSD diagram of ln (population density) versus size. Lognormal by size distributions have been proposed for igneous and metamorphic rocks and materials that crystallise at low temperatures. Lognormal models are difficult to prove with simple linear frequency histograms or on a classic CSD diagram. Instead, it is better to use a normalized cumulative distribution function diagram, which gives a straight line for lognormal distributions. Similarly, fractal size distributions can best be verified simply by using a bi-logarithmic diagram. Statistical measures can be used to invalidate a null hypothesis, but with much caution for real data, especially those derived from two-dimensional measurements and with a limited range in sizes. These methods are applied to CSDs of plagioclase in three igneous rock samples to illustrate the pitfalls of model fitting. A simple nomenclature is proposed for ideal CSD shapes.

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1. Introduction

In petrology we generally examine rocks so that we may understand the nature of the starting materials and the processes which they have undergone. This inverse problem has no unique solution but the petrogenesis of a rock can be constrained by making observations of the widest possible scope. Chemical and isotopic measurements dominate many current petrological studies, but the rock textures should not be ignored: Crystals change in size, shape, orientation and position during solidification or melting, hence, textures can convey petrologic

* Tel.: +1 418 545 5011x5052; fax: +1 418 545 5012. *E-mail address:* mhiggins@uqac.ca. information. Early textural studies were qualitative and did not add much to the petrologic story. However, recent advances in the quantification of textures have considerably augmented their utility. By far the most commonly applied quantitative textural measurement is crystal size distributions (CSD).

The crystal size distribution of a rock is just the number of crystals of a mineral per unit volume within a series of defined size intervals. If a large number of crystals are measured then the width of the size intervals can be decreased and the CSD eventually becomes a smooth function. The CSD is an intrinsic property of a solid, like the density or the composition. Like other parameters, it can be measured and presented in different ways, but there is only one CSD for each

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phase in a rock. A small caveat is perhaps the meaning of crystal size: it can be defined in many ways, but all reflect in some way the dimensions of the crystals (Higgins, 2000). CSDs can vary enormously between different rocks and phases, although they have closure limits, like chemical compositions and other parameters (Higgins, 2002a).

The CSD of rock is generally just a view of the last stage of textural evolution, although glimpses of earlier textures may be sometimes observed (Higgins, 1998). The initial solidification of an igneous rock is dominated by the kinetic effects of crystal nucleation and growth (Fig. 1). However, once crystals have formed then their abundance may be changed by mechanical processes such as sorting and compaction. In many rocks a relaxation of the crystallisation driving force may enable the texture to evolve towards equilibrium, which can be observed in CSDs by coarsening (Higgins, 1998). Each of these processes produces characteristic changes in the CSDs, some of which may converge from different original CSDs towards a unique final CSD shape. Many authors have tried to establish petrologic models from observations of CSD shapes. Most rocks have complex histories and it is clearly necessary to find special situations where one process dominates, or overwrites earlier textures. Marsh (1988) suggested that many igneous CSDs are straight lines when plotted on a graph of ln (population density) versus size. Indeed, this diagram is commonly referred to as the 'CSD diagram'. Some authors have considered that many CSDs have a lognormal frequency distribution (e.g. Eberl et al., 1998; Kile et al., 2000). Others have found CSDs that have a fractal (power law) size distribution (e.g. Armienti and Tarquini, 2002). In this paper I will show how these proposed CSD models can be verified using diagrams and statistical methods that are appropriate for these purposes. I want to also point out the pitfalls associated



Fig. 1. Development of igneous rock textures—role of kinetic, mechanical and equilibrium processes.

with data produced by certain analytical methods and the limitations of model fitting.

2. Theoretical CSDs and appropriate diagrams

Randolph and Larson (1971) developed a theoretical treatment of CSDs in continuously fed, steady-state, industrial crystallizers. They found that growth rates were independent of crystal size and that the CSDs were linear when plotted on a graph of ln (population density) versus crystal size (Fig. 2A). The slope of this graph is equal to -1/(growth rate × residence time). This model is fundamentally kinetic-sorting and equilibration have not occurred. Marsh (1988) proposed that a volcanic magma chamber could approximate such a reactor. Cashman and Marsh (1988) then applied these ideas to a lava lake and determined some of the kinetic controls on crystallisation. However, although a lava lake is not a steady-state system Marsh (1998) concluded that closed systems of batch crystallisation at may also be linear in these coordinates. However, in batch systems the significance of the residence time is not clear. Since then many other studies have found CSDs with similar shapes, now commonly referred to as straight CSDs. Commonly, the residence time has been calculated from the slope and growth rate estimates, even if the system is far from steady-state. Not all authors have adopted this semi-logarithmic CSD model.

Normal or lognormal distributions are often thought to be common in natural systems. An example well known to geologists is chemical abundance, although recent analysis shows that this is rarely true in practice (Reimann and Filzmoser, 2000). Lognormal by volume or mass distributions (i.e. the dependant variable that is lognormally distributed is the mass or volume of material in each size interval, not the numbers of grains) are commonly proposed for sediments (Lewis and McConchie, 1994). A lognormal by number distribution has also been proposed for CSDs of crystals grown at low temperatures in aqueous systems (Eberl et al., 1998; Kile et al., 2000). These authors considered that such lognormal distributions were fundamentally kinetic in origin, as a result of nucleation and growth. They showed that lognormal CSDs can be produced if it is assumed that growth rate is proportional to crystal size. This model has been subsequently extended to magmatic systems, with the assumption that such CSDs are kinetic in origin (Bindeman, 2003). Many metamorphic (Cashman and Ferry, 1988) and plutonic rocks (Higgins, 1998) have CSDs that are concave-down on a semilogarithmic CSD diagram and commonly approximately lognormal. Kinetic effects are clearly important in these



Fig. 2. Graphical display of three different theoretical CSD models on three different diagrams: A) Semi-logarithmic 'Classic' CSD diagram (following March 1988). B) Cumulative Distribution Function (CDF) diagram. C) Bi-logarithmic cumulative size diagram. A 'straight' CSD (CSD-S) has a semi-logarithmic distribution and is straight in diagram A. A lognormal distribution (CSD-L) is straight in diagram B. A fractal distribution (CSD-F) is straight in diagram C.

rocks, however during magma or fluid present conditions the texture may start to equilibrate and coarsen. Therefore, lognormal CSDs in igneous rocks have been interpreted as due to both kinetic and equilibrium effects (Fig. 1).

Clear evidence of lognormal behaviour of CSDs has not always been provided. Some authors have used linear frequency histograms to show that their CSD data are lognormal (e.g. Eberl et al., 1998). However, this is not the best solution, as deviations are not clearly shown, especially for sizes smaller than the mean. A better approach is to use the transform of the cumulative distribution function against ln (size), the CDF diagram, for identifying such distributions, as is done in sedimentology (Fig. 2B; Carver, 1971). If a distribution is lognormal then this graph is a straight line. Statistical verifications of lognormal behaviour are discussed below.

The mathematical concept of fractional dimensions– fractals–is relatively recent (Mandelbrot, 1982), but has had a remarkable impact on the natural sciences, particularly in geology (Turcotte, 1992). Indeed, it is now commonly assumed that most geological structures, including rock textures, are fundamentally fractal. Fractals may be applicable to some phenomena in geology, but it must be proved that they can describe the fundamental component of CSDs. So far there have been few studies in which CSDs have been found to have a fractal distribution: Armienti and Tarquini (2002) found that olivine in the mantle had a fractal distribution. Here, this clearly reflects the balance between grain size reduction caused by deformation and a return to equilibrium via coarsening. In addition Turcotte (1992) found grain sizes of fragmental rocks to be fractal over significant ranges in size. It will be shown below that some purely magmatic CSDs may also be fractal.

The classic CSD diagram was not designed to examine if a CSD is fractal. However, it is easy to construct the CSD diagram for this purpose, such as ln (number of crystals>size) versus ln (size) (Fig. 2C). The fractal dimension of this size distribution is equal to -1 multiplied by the slope (Turcotte, 1992). This will be referred to as a 'fractal diagram'. It is very important that the dataset has a wide range of crystal sizes, otherwise such a fractal behaviour cannot be identified (Pickering et al., 1995). If a fractal diagram has two or more clearly defined slopes for different sizes then it is described as multifractal. However, other distributions are curved on a fractal diagram and some authors may be tempted to

conclude that they are multifractal. It is rare that crystal size distribution data cover a sufficient size range and are sufficiently accurate that multifractal behaviour can be clearly recognised.

More complex distributions can be readily proposed, such as Weibull and Rosin-Rammler (Kotov and Berendsen, 2002) but their utility is probably limited. Many CSDs are the result of several sequential processes, commonly kinetic, mechanical and equilibrium, and hence can have very complex distributions (Fig. 1). A better approach may be to define the effect of different processes on crystals of different sizes and combine these processes stochastically (e.g. Higgins, 1998).

It may be useful to classify the different ideal CSD shapes so that confusing terms such as 'straight CSD' may be avoided. It is proposed here that semilogarithmic CSDs, those that are straight on a classic CSD graph are referred to as S-CSD (Fig. 2). Similarly, lognormal CSDs are L-CSD and fractal CSDs are F-CSD. CSDs that can be produced by a combination of simple CSDs could be designated by two letters. For instance, mixing of two S-CSDs produces a characteristic CSD with two straight segments (Higgins, 1996b) that could be referred to as SS-CSD.

3. Statistical verification of distributions

"If your experiment needs statistics, you ought to have done a better experiment." Ernest Rutherford.

The use of statistical parameters to prop up dubious arguments has a long history (Huff, 1954). Nevertheless, such methods are another approach to verify if a CSD follows an ideal distribution model. This is generally done using the 'null hypothesis': this is the hypothesis that the distribution of the data was produced by random processes (Swan and Sandilands, 1995). If the null hypothesis is validated then the CSD does not follow the model; if it is rejected then it is still possible that the CSD follows the model. The null hypothesis must be tested using a test statistic. There is a level of significance associated with this hypothesis, commonly 5%. This means that there is only a 5% chance that the distribution seen occurred by random processes.

The most widely applicable test uses the value of chisquared.

$$\chi^2 = \sum \frac{(O_j - E_j)^2}{E_j}$$

where *j* is the class interval number, O_j is the observed frequency of that class and E_j is the expected frequency of that class. It is important to note that this parameter is

calculated from the entire distribution. Each class should have at least 5 observations. Classes should be symmetrically disposed around the mean. The significant values of this parameter depend on the number of observations and the required significance level. These values are available in standard statistical texts (e.g. Swan and Sandilands, 1995) or can be calculated using the inverse gamma function.

The chi-squared test is very susceptible to the number of classes (Swan and Sandilands, 1995): The null hypothesis may be more commonly accepted for larger numbers of classes. For a small number of classes failure to reject the null hypothesis (i.e. if we accept that the data conform to the model distribution) may be due to broad and unimportant similarities between the data and the model (Swan and Sandilands, 1995). These deficiencies are generally evident on appropriate CSD graph.

There is another problem with this approach: the value of chi-squared in this simple calculation only indicates the dispersion of the points about the line. It does not say if this dispersion exceeds the individual uncertainty (error) in each point (Fig. 3). A much better assessment of the level of significance can be made if the uncertainty associated with each point is known (Bevington and Robinson, 2003). A goodness of fit parameter, Q, can then be calculated that determines the probability that the observed discrepancies between the observed and expected values are due to chance fluctuations. A very low value indicates that the discrepancies are unlikely to be due to chance fluctuations. This means that either the model is wrong or the



Fig. 3. Goodness of fit of data to a straight line. In these two sets of data, A and B, the distributions of the data points are identical, but data in set B are more precise. The value of chi-squared and the correlation coefficient (r^2) are only derived from the data values and hence are identical for these two data sets. However, in A a line can be drawn through the error bars and the *Q* value is equal to 0.1, both indicating that the null hypothesis can be rejected and the data may follow the distribution indicated by the straight line. In B the same data points have greater precision. A line cannot be drawn through all of the error bars and the *Q* value is less than 0.1, hence the null hypothesis must be accepted and the data do not follow the distribution model.

uncertainties have been underestimated or the uncertainties are not normally distributed. If Q is greater than 0.1 then the null hypothesis is rejected and the CSD may follow the model distribution. A value of Q > 0.001 may be acceptable, because truly wrong models generally have very small values of Q. Values higher than 0.1 do not indicate that the data have more validity. A value close to 1 may indicate that the errors have been overestimated.

The Smirnov–Kolmogorov test is one of the other statistical tests that are commonly used (Swan and Sandilands, 1995). This test is based on the cumulative frequency distribution of the data. The maximum distance between the data and the ideal distribution is evaluated in terms of the percentile. It is most sensitive for the central part of the distribution and least sensitive for very small or large crystals. Unfortunately, it is the number of crystals in these parts of the CSD that show which ideal distribution is most closely followed. Hence, it is probably not very useful for this purpose.

A number of graphs give a straight line if the CSD follows the theoretical distribution. In this case the data values can be regressed by minimizing the value of chi-squared. The value of chi-squared or the correlation coefficient r^2 are commonly used to quantify how closely the observed data correspond to a straight line. However, if the uncertainty in each point is known then the goodness of fit parameter Q can be calculated as above and this is a better measure of the validity of the model. This calculation is done in some statistical programs and CSD reduction programs (e.g. CSDCorrections 1.3).

4. Measurement of real CSDs

A CSD is a physical parameter of a rock that must be measured, wherein lie a number of limitations and sources of uncertainty. Ideally, the most accurate CSD is determined from three-dimensional measurements of crystals in a rock. This can be done using X-ray tomography, serial sectioning, and natural, chemical or mechanical separation of crystals. However, these methods are not commonly used for various reasons: touching crystals are not easy to distinguish in X-ray tomography or serial sectioning and physical crystal separation methods are not widely applicable. In addition the range of crystal sizes that can be measured is limited by the resolution of the method and the maximum image size. Hence, most CSDs are determined from measurements of crystal outlines in sections. If the data are derived from measurements of photographs or digital images then the range of crystal

sizes that can be measured may be restricted. This can be overcome by the combining measurements at two different scales (e.g. Higgins and Roberge, 2003).

It is not simple to calculate a CSD, which is a threedimensional parameter, from two-dimensional data (Peterson, 1996; Higgins, 2000). Indeed there are no exact solutions to this problem (except for spheres) but approximate results can be obtained if the crystal shapes and the nature of the fabric are defined (Higgins, 2000). Conversion cannot be done for individual crystals, but only for sub-populations groups in different size bins. Hence an original continuous 2D distribution becomes discontinuous in 3D. It should be remembered that in many early papers, and a few recent ones, the authors used incorrect conversion equations that resulted in inaccurate, though precise, CSDs. Such data should be converted using more accurate methods (Higgins, 2000).

The nature and position of the lower and upper size limits must be established. One consequence of stereologically correct conversions is that many CSDs have poor precision for small crystal sizes. In addition, many methods have limits to their resolution—for instance the pixels of a digital image. Therefore we must distinguish clearly between a methodological (analytical) lower size limit, where we have no idea if there are smaller crystals, and an actual lack of small crystals. This is particularly important in the identification of lognormal and fractal CSDs. The upper limit of a CSD is generally not well defined, as crystal numbers are small. However, field observations can commonly identify the maximum crystal size.

5. CSD data—typical samples

Three samples have been chosen to represent the common range of CSDs in magmatic rocks and typical two-dimensional analytical methods. Three were chosen from my studies, because I have complete control on the stereological conversion and can also discuss the data quality and limits. Q values are calculated for semilogarithmic (classic) and fractal diagrams as uncertainties in the data points have been calculated. However, this cannot be done for CDF diagrams as the uncertainly in each point is not easy to determine.

Egmont volcano, on Mt Taranaki, New Zealand is an active andesite volcano. The initial study of the plagioclase CSDs of these rocks used an inappropriate conversion method (Higgins, 1996a), but the data were recalculated in a later paper (Higgins, 2002a). Plagioclase is the major phenocryst phase in these rocks, and comprises 7 to 30% of the rock. The sample shown here, NZ-12, has 7% plagioclase (Fig. 4A). The lower size limit of the analytical method was 0.05 mm. The CSD is generally almost straight on a classic CSD diagram (Fig. 5A). The CSD descends for the lowest size interval, but this probably reflects the error in that point. The goodness of fit, Q, of the CSD to a straight line found to be 0.42, which is greater than the value of 0.1 needed to indicate significance (these calculations were done by the program CSDCorrections version 1.36, Higgins, 2000). Hence, we can surmise that the CSD is linear on a classic CSD diagram and describe it as an S-CSD.

The same data also give a straight line when plotted on a CDF diagram (Fig. 5B). This might be taken as proof that the data are lognormal, which conflicts which the conclusion above. The problem here is that the data are undefined below a size of 0.05 mm. We do not know from the CSD what happens below this point. If the CSD were lognormal then there would be few or no microlites (Fig. 2B). Inspection of the thin section shows that there are, indeed, microlites and hence that the CSD does not terminate at this size and therefore the CSD cannot be lognormal (Higgins, 1996a). In an ideal situation this would be verified by more detailed analysis of small crystals. Here, however, I wish to illustrate the problem with data typical of many studies.

These CSD data are strongly curved on a fractal diagram (Fig. 5C). No three points are collinear as would be expected if the texture were fractal. The Q value is 2.2×10^{-18} , clearly indicating that it is far from linear.

Soufriere Hills volcano, Montserrat, is also an andesite volcano that started its most recent eruption in 1996 (Robertson et al., 2000). Plagioclase is the major phase and has a large range in size from microlites to phenocrysts 1.5 mm long (Fig. 4B; Higgins and Roberge, 2003). For this reason, intersection data were collected at two different scales and combined to give a single CSD with a much greater range in measured crystal sizes than the Egmont volcano sample (Fig. 5A and D). Inspection of thin sections confirms that crystals continue below the smallest size measured in this study (0.04 mm), hence the CSD is undefined below this size. On a semi-logarithmic diagram the CSD is strongly curved concave-up throughout the measured range in crystal sizes (Fig. 5D). This is reflected in the Q value of 1.18×10^{-43}

On a CDF diagram (Fig. 5E) almost the entire curve lies above the median size (0.045 mm). This is because about half of the small crystals are in the lowest size bin. The CDF is strongly curved also, firmly establishing that this distribution is not lognormal, even if the number of crystals smaller than the detection limit is not known.



Fig. 4. Digitized outlines of the three thin sections that were used to construct the CSD diagrams of Fig. 5. A: NZ 12, plagioclase in an andesite lava from Egmont volcano, Mt Taranaki, New Zealand (Higgins, 1996a). These data were recalculated using CSDCorrections 1.36 (Higgins, 2000). B: MMMon2, plagioclase in an andesite from Soufriere Hills volcano, Montserrat (Higgins and Roberge, 2003). The section was digitized at two different scales, so that a larger size range of crystals could be quantified. C: MH-94-60, plagioclase in troctolite from the Kiglapait intrusion, Nain, Canada (Higgins, 2002b).



Fig. 5. Three contrasting rock CSDs displayed in three different ways: A, D, G) Classic CSD diagram; B, E, H) Cumulative distribution function (CDF) diagram; C, F, I) bi-logarithmic diagram. Crystal size is in mm. The uncertainly in the points on the classic CSD diagram is that calculated by the program CSDCorrections (Higgins, 2000). These uncertainties are also used in the fractal diagram. Uncertainties in points on the CDF diagram can only be calculated for 3D measurements and are hence absent here. The nature of each data point is shown in the legend at the bottom. A vertical line indicates a true termination of the CSD and that there are no smaller or larger crystals. If a CSD ends in an open circle then the CSD is not defined for smaller or more rarely larger crystals. This type of termination is an artefact of the analytical method.

The CSD is almost straight on a fractal diagram (Fig. 5F) with a fractal dimension of 2.4. It should be noted that the range in sizes here is nearly two orders of magnitude, much greater than that of the other samples discussed here. However, the Q value for all data is 4×10^{-22} , which reflects the deviation from the trend of the leftmost point. It is likely that the error in this point has been underestimated: If it is omitted then Q=0.04 and the CSD may be considered to be fractal and termed F-CSD.

The *Kiglapait intrusion, Labrador, Canada* presents one of the best-exposed sections through a mediumsized layered gabbroic intrusion (Morse, 1969). I have recently published CSDs of plagioclase, olivine and clinopyroxene (Higgins, 2002b). The sample shown in Fig. 4C differs considerably from the two samples discussed earlier as it is completely crystalline and all crystals have been measured. The plagioclase CSD shown in Fig. 4D is typical of the intrusion. This plutonic rock differs from the volcanic samples in that it The right part of the CDF diagram is almost straight, as for the Taranaki sample, but again almost all the data lie to the right of the mean (Fig. 5H). In addition, the left end of the CDF descends vertically to - infinity, as there are no small crystals. Overall, this is very far from a lognormal distribution.

The CSD on the fractal diagram (Fig. 5I) is strongly curved, again indicating a lack of self-similarity. At small sizes the curve becomes horizontal as no more crystals are added when the size is decreased. Hence, the CSD is clearly not fractal.

6. Conclusions

Different diagrams can be used to verify if a CSD dominantly follows a known distribution model. A semi-logarithmic distribution, as proposed originally for rocks by Marsh (1988), can be verified using the classic CSD diagram of ln (population density) versus size. However, attention must be paid to the nature of the lower size limit: measurement artefact or real size limit. Few samples follow this model exactly, but many basic volcanic rocks have almost straight CSDs on this diagram.

Lognormal distributions are particularly difficult to prove, partly because of the poor precision of data on smaller crystals. A good approach is with the CDF diagram. However, again the nature of the lower size limit is very important. Semi-logarithmic distributions can closely resemble lognormal distributions except that they do not lack small crystals. It may be necessary to combine methods or measure the data at two different scales to prove this distribution. If a CSD is lognormal then we would commonly call the texture porphyritic.

Fractal size distributions can be verified using a bilogarithmic diagram. Again, a large range in measured crystal sizes is commonly necessary. If a CSD is fractal, then we would commonly call the texture bimodal or even porphyritic. The term bimodal is a confusing one as there is clearly only one size mode. What is generally meant is that there are two populations of crystals. Clearly, this texture is completely different from a texture with a lognormal CSD, which was also described as porphyritic, as it contains small crystals. However, this distinction is not always made by petrographers.

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Appendix A. Calculation methods

Calculation of the 'Classic CSD Diagram' is described in many studies (e.g. Higgins, 2000). In brief, the vertical axis is the natural logarithm of the population density. The population density is the number of crystals divided by the volume and the width of the size interval measured. The horizontal axis is the size, generally interpreted as the longest 3D dimension of the crystal. The Q value can be calculated using CSDCorrections 1.36 or statistical programs, but not with most general purpose spread-sheet programs.

The CDF diagram is explained in many texts on sedimentary petrology (e.g. Carver, 1971). The vertical axis is derived from crystal numbers in each interval. For 3D data-sets this is easily determined, but is more complex for section data. CSDCorrections 1.3 (Higgins, 2000) gives the crystal number in each interval, which can be used to calculate this parameter. The crystal numbers are summed and normalized so that the largest interval has a value of 1 and the smallest interval is 0. These values give the familiar double curve of a normal distribution. They can be linearised using the inverse function of the standard normal cumulative distribution (Available in Excel as the function NORMSINV). The horizontal axis is the natural logarithm of the size, but base 10 logarithms can also be used. There are a number of dedicated programs that can be used to fit lognormal and other distributions to size data. One of the more widely used is STF (sequential fragmentation/transport) by Ken Wohletz (http://www.ees1.lanl.gov/Wohletz/ SFT.htm).

Many different diagrams can be used to determine if a size distribution is scale invariant (Turcotte, 1992). Here, the vertical axis is the natural logarithm of the number of crystals larger than the true, 3D size. The horizontal axis is the natural logarithm of the size. Natural logarithms are used here so that the data more resembles those in the other diagrams, but base 10 logarithms can also be used for both axes. It should be noted that aspects of the texture other than the size may also be tested for scale invariance, such as the actual space occupied by the crystals or their outline.

An Excel spreadsheet is available at http://wwwdsa. uqac.ca/%7Emhiggins/csd_calc.xls that shows sample calculations.

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