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# A new segregation index for solid multicomponent mixtures

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#### Abstract

Several indices can be found in the literature in order to quantify the mixing degree of two component mixtures in fluidized beds, but none of them is actually capable of describing how a specific component of the mixture is distributed. Many of these indices may be influenced by the experimental procedure used for evaluating the mixture, such as the number of vacuumed layers, or equivalently the layer thickness, since the solids distribution is generally measured in layers (or at most in cells) and not in a continuous way along the bed. In the present work, a novel set of indices for studying segregation is proposed: the Three Thirds Segregation Indices Set, is developed allowing to characterize not only the segregation level, but also the segregation pattern of a specific component of interest. The set is also compared, tested and validated with other existing indices (M index by Rowe et al.)(1972) and "s" index by Goldscmidt et al. (2003)), and experimentally verified. As a result of these tests, it is found that M and "s" indices do not allow to compare experiments performed with different numbers of layers, and they do not distinguish different segregation profiles leading to some mistakes when in the proximity to the extreme cases (Full Central or Full Bottom segregation). On the opposite, the new set of indices turns out to be independent on the number of layers, and to minimize experimental errors or the discrepancies caused by applying different experimental procedures.

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#### 1. Introduction

Mixtures of solids are widely used in the process industry (such as food, petrochemical, or cement industries). According to the process characteristics and the desired final product, mixing can be carried out in different types of devices, such as mixers or fluidized beds. However, as solids mixtures are usually composed of particles differing in density and/or size, they may segregate or do not mix properly.

During the 1960s and 1970s a considerable number of indices were developed in order to quantify and characterize the mixing or segregation of a binary mixture and the performance of different types of mixers, such as the static mixer, the conical rotating mixer, the ribbon mixer, or the twin shell mixer, used mainly in the cement industry. As the evaluation of the mixture quality in these devices is performed by sampling, many of these indices were based on statistics; perfectly ordered, randomly mixed, or totally segregated mixture are then generally considered as limit cases [1, 2, 3, 4].

Mixing performances are also important in other types of devices, such as fluidized beds or spout-fluid beds, often employed in the process industry, as chemical reactors or dryers; the atmospheric lyophilization of food by immersion in adsorbent material is an example of a complex binary mixture for which segregation can impair dramatically performances. In the fluidized bed air is passed through a perforated plate, while in the spout-fluid bed air is injected by means of a main injector (like a spouted bed) and lateral injectors, conferring a better mixing performance to the apparatus.

Many segregation or mixing indices were also developed specifically for characterizing mixing and segregation in fluidized beds, and some of them are based on statistical concepts similarly to those used for mixers. Di Renzo et al [5] applied the *Mixing Index* proposed by Lacey [6], involving the variance of the concentration distribution in the particle system, to analyze segregation in a binary system of particles with different density and equal size. Zhang et al [7] utilized the "Shannon entropy", which is also based on statistics, as indicator of the mixing performance evaluating the dynamics of mixing and the effect of time and particle density. Barghi et al [8] proposed a mixing index based on collisions between aluminum tracer particles and the probes, assuming that the collisional frequency is proportional to the concentration of particles in a given bed height. Methods to predict the segregation behaviour of a mixture from the particle and fluid physical properties have also been proposed: Escudié et al [9] used a "reduced bulk density" (taking into account particles and fluid densities) as indicator of the segregation degree in a liquid fluidized bed.

Among the segregation indices, one of the most widely used is the M mixing index proposed by Rowe et al. [10], based on jetsam fractions in the upper part  $(X_J)$  and the whole of the bed  $(\overline{X}_J)$ :

$$M = \frac{X_J}{\overline{X}_J} \tag{1}$$

This index can be applied as long as the jetsam and flotsam are clearly identified, bottom segregation is preponderantly dominant, and jetsam concentration and particle size are such that eventually one layer whose concentration is around 100% may be found. Thus, M = 1 corresponds to perfect mixing, whereas M = 0 means complete segregation.

In addition, Wu and Baeyens [11] reviewed predictive equations for this index, and for mixture minimum fluidization velocity calculation available in literature, and proposed a new equation for predicting M mixing index based on their experimental results.

On the other hand, Goldschmidt et al. [12] developed a segregation index suitable for digital image analysis of segregation in fluidized beds, the "s" index, which takes into account the composition along all the bed of both components of a binary mixture, and normalizes it considering a theoretical maximum segregation degree of the mixture. The mathematical formulation of the index is the following:

$$s = \frac{S-1}{S_{\max}-1} \tag{2}$$

where S is the ratio of an average layer height of small particles to the same quantity calculated for large particles,

$$S = \frac{\langle h_{\rm small} \rangle}{\langle h_{\rm large} \rangle} \tag{3}$$

and  $S_{\text{max}}$  represents the theoretical maximum degree of segregation,

$$S_{\max} = \frac{2 - x_{\text{small}}}{1 - x_{\text{small}}} \tag{4}$$

where  $x_{\text{small}}$  is the overall mass fraction of small particles in the bed.

Moreover, in their work the authors used particles of two different sizes colored according to their diameter. Therefore, after taking images during the fluidization and dividing the generated pictures in cells, they analyzed the color distribution, and calculated the solid volume fractions in each cell from the total area of pixels identified as particles in each cell. Thus, the numerator and denominator of Equation 3 were calculated as follows:

$$\langle h_{\text{large}} \rangle = \frac{\sum_{k} x_{\text{large}} \alpha_{\text{large},k} h_k V_k}{\sum_{k} x_{\text{large}} \alpha_{\text{large},k} V_k} \tag{5}$$

$$\langle h_{\rm small} \rangle = \frac{\sum_k x_{\rm small} \alpha_{\rm small,k} h_k V_k}{\sum_k x_{\rm small} \alpha_{\rm small,k} V_k} \tag{6}$$

where  $\alpha_{\text{large},k}$  or  $\alpha_{\text{small},k}$  is the total volume fraction of small or large diameter particles in the cell (depending on the case),  $x_{\text{large}}$  or  $x_{\text{small}}$  is the overall mass fraction of large or small particles in the bed, and  $h_k$  and  $V_k$  represent, respectively, the height of the center of the cell k from the air distributor and the cell volume.

Therefore, a value of 1 for the "s" index corresponds to a completely segregated system, whereas s=0 means perfect mixing. This index can be also extended to ternary mixtures as done by Olaofe et al [13] for mixtures composed of glass particles of equal density and different diameter.

Despite several indices were proposed by different authors in order to quantify the segregation level or mixing of a binary mixture in a fluidized bed, none of them is actually able to describe how a specific component of the mixture is distributed along the bed. In other words, sometimes it is important not only to know how much the binary system differs from the uniformity, but also the distribution of a certain component of interest.

On the other hand, proposed indices are generally influenced by the experimental procedure used for evaluating the mixture, such as the number of vacuumed layers (or the layer thickness), since the solids distribution is measured in layers (or at most in cells) and not in a continuous way along the bed. Consequently, it might be difficult to compare experiments done with non-equal number of layers and in fluidized beds of different size, and generally to compare results presented by different authors.

As it was mentioned above, the previously proposed segregation indices do not describe the shape of the segregation profile. In addition, according to the results of the theoretical tests hereafter described, they sometimes do not give completely accurate results, in particular when central segregation is present or the number of experimental layers varies even for the same segregation pattern.

Therefore, the main scope of the present work is to develop an appropriate segregation index which allows the comparison among different experiments independently of the equipment details and the experimental procedure used. It will be tested both with simulated and experimental cases and evaluated in comparison with other existing indices (M and "s").

#### 2. The new segregation index

A new set of indices is proposed, based on the measure of the distribution of the material of interest along the bed and the segregation level; this differs from previous indices which use only one quantity to evaluate the segregation. This set is denominated Three Thirds Segregation Set of Indices (TTSIS), and is defined as:

$$TTSIS = [p_{\rm I}, p_{\rm M}, p_{\rm S}]_{\aleph_2} \tag{7}$$

where  $p_{\rm I}$  is the Bottom Third Indicator,  $p_{\rm M}$  is the Middle Third Indicator,  $p_{\rm S}$  is the Top Third Indicator, and  $\aleph_2$  is the Segregation Level. Defining  $F_q(h^*)$ , the accumulated mass of material of interest "q", as a function of the dimensionless bed height from the bottom  $(h^*)$ , these three indicators are calculated as follows:

$$p_{\rm I} = \frac{F_{\rm q}\left(\frac{1}{3}\right)}{m_{\rm qT}} \tag{8}$$

$$p_{\rm M} = \frac{F_{\rm q}\left(\frac{2}{3}\right) - F_{\rm q}\left(\frac{1}{3}\right)}{m_{\rm qT}} \tag{9}$$

$$p_{\rm S} = \frac{m_{\rm qT} - F_{\rm q}\left(\frac{2}{3}\right)}{m_{\rm qT}} \tag{10}$$

and

$$\aleph_2 = \max(p_{\mathrm{I}}, p_{\mathrm{M}}, p_{\mathrm{S}}) - \min(p_{\mathrm{I}}, p_{\mathrm{M}}, p_{\mathrm{S}})$$
(11)

where  $m_{qT}$  is the total mass of the material of interest in the bed.

The extreme values and their meaning are described in Table 1. Naturally, in the experimental work it is more usual finding intermediate distribution patterns rather than these extreme situations. Table 2 shows the adopted criteria for classifying the intermediate cases, establishing the numerical bands for each indicator. Moreover, Figure 1 schematically represents the particle distribution for the extreme cases and intermediate situations.

During the experimental work, this criterion was implemented by means of a computational code in Python and the classification of each experimental case was automatically obtained in the course of the results post-processing stage. The bands adopted for the classification of the different segregation types were fixed on the basis of a preliminary experimental campaign. It must be evidenced that to have a rapid idea about the distribution of the product of interest along the bed and the segregation degree, it is sufficient to consider the Segregation Level ( $\aleph_2$ ) and the Segregation Type. Otherwise, if the influence of a certain variable on the segregation profile is of interest, the variation of the whole set of indices should be taken into account.

The most important advantage of the TTSIS is that it quickly gives an intuitive idea about the segregation profile, capturing somehow the shape of the distribution curve, and is simple to be calculated. However, its main disadvantage is the fact that the volume occupied by the product of interest cannot be greater than one third of the bed volume, to avoid incoherent results (like a TTSIS of the form  $[p_{\rm I} > 0, p_{\rm M} > 0, p_{\rm S} < 1]$  for a full top segregation).

#### 3. Materials and Methods

#### 3.1. Theoretical test of segregation indices

When segregation experiments are carried out in a fluidized bed with a binary mixture, in order to determine the distribution of a solid of interest along the bed, the mixture must be fractionated. One technique is by dividing the bed into layers, vacuuming and sieving each one. However, in this way, all the factors related with the experimental procedure and apparatus may influence the results of the segregation indices. For example, sometimes the height of the layer,  $\Delta h_i$ , cannot be directly measured with accuracy as a consequence of the configuration of the apparatus or other causes, and can be determined as,

$$\frac{m_{\rm si}}{m_{\rm sT}} = \frac{\Delta h_i}{h_{\rm bed}} \tag{12}$$

that is from the ratio of mass in the vacuumed layer,  $m_{\rm si}$ , and the total mass of the solid in the bed,  $m_{\rm sT}$ .

Assuming a bed with j components, this estimation is valid provided that the solids bulk density of each layer i is equal to the overall solids bulk density in the bed, that is,

$$\sum_{j} \alpha_{ji} \rho_j = \sum_{j} \alpha_{jT} \rho_j , \, \forall i$$
(13)

With the objective of investigating the influence on segregation indices of two experimental factors, the number of layers and the layer actual height, and the effect of eventual differences among the solids bulk density of each vacuumed layer, a mathematical test was performed, considering an hypothetical fluidized bed containing a mixture of 12 kg of bran and 87.8 g of lyophilized carrot discs. Four kinds of segregation patterns were considered, distributing the food material in different ways along the bed (Full Top, Full Central, Full Bottom, and Pure V), and all the other variables were calculated, considering not only the mass distribution of bran and carrots and their volumetric fractions, but also the space occupied by air (void fraction). Then, number of layers, layer thickness, and solids density were varied, and mass balances were solved for each case in order to obtain a coherent mass distribution. For full top as well as full bottom segregation, it was imposed that all the food material concentrates in the upper and lower layers respectively, whereas for central segregation, a concentration was set in the two middle layers. For V-segregation, it was assumed that one half of the mass of carrots concentrates in the top layer, and the other half in the bottom one. Regarding the fluidized bed, a volume of  $0.0482 \text{ m}^3$  and a sectional area of  $0.1225 \text{ m}^2$  were assumed.

A mass distribution vector  $(\vec{P}_{\text{dis}})$ , of dimension  $n_{\text{lyr}}$  ( $n_{\text{lyr}}$ : number of layers), was defined in order to impose the product distribution along the bed. The value of each component of  $\vec{P}_{\text{dis}}$  was varied between 0 and 1, according to the type of segregation considered. Then, the mass of product in each layer,  $m_{\text{P}i}$ , is calculated with the following expression,

$$m_{\rm Pi} = m_{\rm PT} P_{\rm dis,i} \tag{14}$$

where  $m_{\rm PT}$  is the total mass of product (87.8 g of lyophilized carrot discs in the considered example).

Depending on the type of segregation profile evaluated, the components of the mass distribution vector,  $P_{\text{dis},i}$ , were fixed as shown in Table 3.

The overall bulk density of the solid phase,  $\rho_{sT}$ , is

$$\alpha_{\rm sT}\rho_{\rm sT} = \alpha_{\rm AT}\rho_{\rm A} + \alpha_{\rm PT}\rho_{\rm P} \tag{15}$$

where, said  $V_{\rm T}$  the bed volume and  $\alpha_{\rm sT}$  the total volume fraction of solids,

$$\alpha_{\rm AT} = \frac{m_{\rm AT}}{\rho_{\rm A} V_{\rm T}} \quad and \quad \alpha_{\rm PT} = \frac{m_{\rm PT}}{\rho_{\rm P} V_{\rm T}} \tag{16}$$

Depending on the factor studied, the mass of adsorbent and the volume of each layer were calculated applying the equations described in the following subsections. Also, as it can be noted, the subscript A (for adsorbent) is used in all variables related with bran, whereas the subscript P (for product) is used in all variables related with lyophilized carrot.

#### Influence of the number of layers

The mathematical test was performed assuming 2, 6, 10 and 100 vacuumed layers. Despite the last value is not realistic, it was considered for evaluating the behaviour in the limit case of infinite layers.

When only the effect of the number of hypothetical vacuumed layers was evaluated, that is, ideal cases without variations in the solids bulk density along the bed and layers with equal thickness, the layer volume  $(V_i)$ , the total mass of solids  $(m_{si})$  and of adsorbent in each layer  $(m_{Ai})$ , were calculated as follows:

$$V_i = \frac{V_{\rm T}}{n_{\rm lyr}} \tag{17}$$

$$m_{si} = \rho_{\rm sT} \alpha_{\rm sT} V_i \tag{18}$$

$$m_{\mathrm{A}i} = m_{\mathrm{T}i} - m_{\mathrm{P}i} \tag{19}$$

#### Non-uniform distribution of solids bulk density

As the solids bulk density of each layer depends on the volume fractions of the mixture components present in each layer, the use of Equation 12 would introduce an error in the estimation of the layer thickness, and consequently, an error on the segregation index calculation. Thus, the effect on segregation indices of a non-uniform solids bulk density distribution was evaluated by varying the solids bulk density of each layer. These variations were imposed through the following steps:

1. Determination of a density variation factor for each layer  $(k_{co,i})$ , defined as:

$$k_{\mathrm{co},i} = \frac{\alpha_{\mathrm{si}}\rho_{\mathrm{si}}}{\alpha_{\mathrm{sT}}\rho_{\mathrm{sT}}} \tag{20}$$

- 2. According to  $k_{co,i}$ , estimation of a first approximation for the adsorbent volume fraction and its mass for each layer.
- 3. Correction of the mass of adsorbent in some layers, in order to keep the given mass of adsorbent in the whole bed (12 kg in this case).

Therefore, in the first step, the density variation factor for each layer was calculated by means of two straight lines with positive and negative slope intercepting in a maximum value at the bed center (Figure 2), assuming that if there was a variation of the solids bulk density, its maximum would be located in the central layers,  $c_e$ .

Moreover, the  $k_{co,i}$  maximum value was imposed for the test  $(k_{co,max})$ , whereas its minimum value was calculated as the reciprocal of this maximum.

Therefore, the solved equations were the following,

$$k_{\rm co,min} = \frac{1}{k_{\rm co,max}} \tag{21}$$

$$c_e = \begin{cases} \frac{n_{\rm lyr}}{2} + 1 & \text{if } n_{\rm lyr} \text{ is even.} \\ \\ \lceil \frac{n_{\rm lyr}}{2} \rceil & \text{if } n_{\rm lyr} \text{ is odd.} \end{cases}$$
(22)

where the symbol  $\lceil x \rceil$  is the ceiling function of x, i.e., the smallest integer not smaller than x.

The slopes of the curves for density variations were obtained with

$$\gamma_1 = \frac{k_{\rm co,max} - k_{\rm co,min}}{c_e - 1} \tag{23}$$

$$\gamma_2 = \frac{k_{\rm co,min} - k_{\rm co,max}}{n_{\rm lyr} - c_e} \tag{24}$$

Thus,

$$k_{\text{co},i} = \begin{cases} \gamma_1(i-1) + k_{\text{co,min}} & i < c_e \\ k_{\text{co,max}} & i = c_e \\ \gamma_2(i-c_e) + k_{\text{co,max}} & i > c_e \end{cases}$$
(25)

In the second step, the first approximation  $(\alpha_{Ai_0})$  for the layer adsorbent volumetric fraction is estimated by

$$\alpha_{\mathrm{A}i_0} = \frac{k_{\mathrm{co},i}\alpha_{\mathrm{sT}}\rho_{\mathrm{sT}} - \alpha_{\mathrm{P}i}\rho_{\mathrm{P}}}{\rho_{\mathrm{A}}} \tag{26}$$

where  $\alpha_{Ai_0}$  is derived by combining Equations 15 and 20. Then, the first approximation for the mass of adsorbent and the total mass of solids in each layer are given as follows:

$$m_{\rm Ai_0} = \alpha_{\rm Ai_0} \rho_{\rm A} V_i \tag{27}$$

$$m_{\mathrm{s}i_0} = m_{\mathrm{A}i_0} + m_{\mathrm{P}i} \tag{28}$$

Since  $k_{co,i}$  is a discrete function of the layer number, the total mass of adsorbent in the bed obtained applying this factor might not be the total mass of adsorbent imposed at the beginning. Consequently, in the third step of this procedure the total mass of adsorbent in the bed is calculated and compared with the original mass. That is,

$$m_{\rm ATc} = \sum_{i} m_{\rm Ai_0} \tag{29}$$

$$m_{\rm A,add} = m_{\rm AT} - m_{\rm ATc} \tag{30}$$

Then if there is an excess, the exceeding quantity is subtracted from the top and bottom layers; on the contrary, the lacking quantity is added to the intermediate layers:

if 
$$m_{A,add} > 0$$
, then :  
 $m_{A(c_e-1)} = m_{A(c_e-1)_0} + 0.5m_{A,add}$   
 $m_{A(c_e+1)} = m_{A(c_e+1)_0} + 0.5m_{A,add}$   
if  $m_{A,add} < 0$ , then :  
 $m_{A1} = m_{A1_0} + 0.5m_{Aadd}$   
 $m_{An_{lyr}} = m_{An_{lyr}} + 0.5m_{A,add}$ 
(31)

Thus, the final values of volumetric fractions of adsorbent, food product, and air can be obtained as follows:

$$\alpha_{\mathrm{P}i} = \frac{m_{\mathrm{P}i}}{\rho_{\mathrm{P}} V_i} \tag{32}$$

$$\alpha_{\mathrm{A}i} = \frac{m_{\mathrm{A}i}}{\rho_{\mathrm{A}} V_i} \tag{33}$$

$$\alpha_{\rm air} = 1 - (\alpha_{\rm Ai} + \alpha_{\rm Pi}) \tag{34}$$

As in these cases a uniform layer thickness was considered, its value was calculated with the following equation:

$$\Delta h_i = \frac{h_{\rm bed}}{n_{\rm lyr}} \tag{35}$$

The values applied for  $k_{\rm co,max}$  in the present test were 1.025 and 1.05.

#### Variation of layer thickness

As the layer thickness and bed height are independent variables directly or indirectly utilized for evaluating the three tested segregation indices (TTSIS, M and "s"), it is important to consider what would be the effect of nonuniform layer thickness on the estimated values of these indices. Therefore, the Top, Central, and Bottom layers thickness was varied by  $\pm 40\%$  with respect to the other ones, whose thickness remained equal. To this purpose the following steps were taken:

- 1. Definition of a primary thickness distribution vector  $(\vec{h}_{\rm dis0})$  with  $n_{\rm lyr}$  components equal to 1.
- 2. Substitute the *n*-th component of  $\vec{h}_{dis0}$  by 0.6 or 1.4 (*n*-th: number of layer to be changed), as explained in Table 4.
- 3. Normalization of  $\vec{h}_{dis0}$  and determination of the final thickness distribution vector  $(\vec{h}_{dis})$ .
- 4. Calculation of the layers thickness and volume by multiplying each element of the thickness distribution vector by the total bed height and volume, respectively.

For the normalization of the primary distribution vector, the sum of its elements was calculated first,

$$S_u = \sum_i h_{\text{dis}0,i} \tag{36}$$

then, the thickness distribution vector was determined as

$$\vec{h}_{\rm dis} = \frac{\dot{h}_{\rm dis0}}{S_u} \tag{37}$$

Therefore, the layer thickness and volume were obtained with the following equations:

$$\Delta h_i = h_{\mathrm{dis},i} h_{\mathrm{bed}} \tag{38}$$

$$V_i = h_{\mathrm{dis},i} V_T \tag{39}$$

Hence, the masses of adsorbent and product, and the foodstuff and adsorbent volume fractions are calculated with the procedure previously described (with  $k_{co,max} = 1$  if density variations were not considered).

#### 3.2. Criteria applied for evaluating the M index

As in the two component mixtures considered in the present work the lighter component, the food product, is also the larger one, it is not possible to establish a priori whether it will tend to sink or float. Therefore, it is not possible to evaluate the M index in terms of jetsam as it was originally defined. However, since the component of interest expected to segregate is the food material and its overall mass fraction is quite low, the M index was not evaluated in terms of concentrations of the jetsam, but in terms of the mass fraction of foodstuff. Anyway, the meaning of the index remains the same. In addition, as "upper" part of the bed it was considered its upper 40%.

#### 3.3. Adaptation of the "s" index

Since the numerator and denominator of Equation 2 (assuming that the solid phase bulk density along the bed is approximately constant) were originally formulated for cell analysis from a grid, they had to be adapted for layer analysis. Working out the original equations (5 and 6), it comes:

$$\langle h_{\text{large}} \rangle = \frac{\sum_{i} m_{\text{P}i} h_i}{m_{\text{PT}}}$$
(40)

$$\langle h_{\rm small} \rangle = \frac{\sum_i m_{\rm Ai} h_i}{m_{\rm AT}} \tag{41}$$

where  $h_i$  is the height from the distributor to the layer center *i*,  $m_{\rm Pi}$  and  $m_{\rm Ai}$  represent, respectively, the mass of food product and adsorbent in the layer *i*, and  $m_{\rm PT}$  and  $m_{\rm AT}$  are the total mass of product and adsorbent in the bed.

#### 3.4. Experimental test

The performance of the segregation indices, in particular the TTSIS, was experimentally tested by means of experiments carried out in the framework of an investigation about atmospheric freeze drying with use of adsorbent. Different two-component mixtures composed of non-food wheat bran and lyophilized, partially lyophlized, or fresh vegetables were used (see Table 5 for more information).

The experiments were performed in a square based fluidized bed (Figure 3(a), 350 mm side), applying different fluidization velocities (0.26, 0.29, and 0.44 m/s), as well as in a spout-fluid bed (Figure 3(b), 200x100 mm), with a single air velocity (0.51 m/s). After a fixed fluidization time (between 20 and 40 min, depending on the case), air was stopped, the bed was divided in layers and the mixture was vacuumed by means of a vacuum machine. Finally, the mixture was sieved and its components were separately weighted. Further details about the apparatus and the experiments can be found in Coletto [14].

Segregation cases from literature were also considered for testing the TTSIS. All of them were carried out using binary mixtures employing different fluidized beds: Wu and Baeyens [11], cylindrical, 300 mm ID.; Qiaoqun et al. [15], rectangular base,  $245 \times 450$  mm; Olivieri et al. [16], cylindrical, 120 mm ID; Rowe et al. [10], cylindrical, 141 mm ID.

#### 4. Results and discussion

Table 6 shows the most representative results of the theoretical tests of the segregation indices.

In Table 6 it can be seen that neither M nor "s" indices are independent of the number of layers used to subdivide the bed. Results obtained applying M present some differences when the number of layers is varied for the same kind of segregation (e.g., for Full Central segregation, M is 0.5 for six layers, while it is 0.0 for 10 and 100 layers). Regarding the "s" index, unsatisfactory results were obtained as well. In fact, not only it does not recognise any difference between Full Central and Pure V segregations, but also its value for both cases is 0.0 (as for the perfectly mixed cases). In addition, "s" index values for Full Bottom segregation show a great dependence on the number of layers. In conclusion, previous results evidence that these indices do not differentiate among the different kinds of segregation, but their estimates are also affected by the experimental procedure followed (in particular by the number of experimental vacuumed layers) making very difficult to compare results of different experiments.

On the other hand, TTSIS present the expected values for each kind of segregation imposed when only the number of layers is varied (only for the limit case where just two layers are considered the predictions are incorrect). In fact, the minimum number of vacuumed layers for analysing segregation applying the TTSIS is three, as this set of indices divides the bed in three parts. Thus, if just two layers are considered, a fictitious middle layer is created when calculating the Middle Third Indicator containing part of the material of interest. Consequently inconsistent values for TTSIS indicators are obtained.

The solids bulk density distribution seems to have slight effect on all the tested segregation indices. For example, considering their values for Full Top or Uniform patterns, M as well as "s" present practically no variations when  $k_{\rm co,max}$  is greater than 1.000. Similarly, none of the TTSIS indices was affected.

When the thickness of the layers is not the same, but a variation is considered in some of them (results in the last five rows of Table 6), TTSIS exhibits some differences with respect to its expected values. Anyway, the deviations from the expected values observed for the M index are considerably greater than those found for TTSIS. For example, for a Full Central segregation pattern, with 6 vacuumed layers, and a change of +40% in the bottom, central, and top layers, M is 0.7 (while it should be 0.0), whereas  $\aleph_2$ of TTSIS gives a difference of at most -16% (between 0.84 and 0.93) against its expected value for this pattern (1.00).

Thus, it can be seen that all tested indices present deviations from their expected values for a given segregation pattern when some parameter is changed. These deviations are a consequence of their mathematical definition. For example, it can be noted that for Full Central segregation, Mindex depends on the number of layers giving different values for six or more layers (Table 6). This discrepancy can be explained with the fact that part of the central layers lies in the upper 40% of the bed whereas when ten layers are considered, the central ones are situated just below this upper 40%. Even though as "upper" part of the layer it was considered the 40% of the bed, similar situations will be found taking a different bed fraction. On the other hand, the dependence on the number of layers for Full Bottom segregation cases shown by "s" index is a consequence of the fact that the denominator of Equation 3 ( $\langle h_{\text{large}} \rangle$ ) decreases when the number of layers is increased. Since in this kind of segregation it was considered that all the product is in the lowest layer, increasing  $n_{\rm lyr}$  the height from the distributor to the bottom layer center  $(h_i, \text{ in Equation 40})$  reduces and the product  $m_{\text{P}i}h_i$  is lower. Moreover, the numerator of Equation 3 remains practically constant due to the great quantity of adsorbent considered, and the theoretical maximum segregation estimated by Equation 4 does not depend on the number of layers. Consequently, the value given by "s" index increases.

Focusing on the experimental cases, in Table 7 are shown the applications of the TTSIS, and M and "s" indices for six experimental samples carried out in a fluidized bed using a two component mixture composed of non-food wheat bran and lyophilised vegetables. In addition, Figure 4 presents experimental values of  $\aleph_2$  obtained for several tests (in the ordinates) compared with the values of M and "s" (in the abscissa). The experiments presented in Figure 4 were carried out in both fluidized bed and spout-fluid bed utilizing vegetables with different level of drying (see Coletto [14] for more information about the mixtures and food characteristics).

In general, it can be observed that M as well as "s" take values not only within their limit interval (between 0 and 1, according to their definition), but also outside it. Even more, it can be noted that a second branch of each curve appears when their limit intervals are crossed; for the  $\aleph_2$  vs. Mcurve, this second branch appears for M > 1, while for the  $\aleph_2$  vs. "s" curve, it appears for s < 0.

Concerning the M index, it exceeds its maximum value (1.00) mainly for Top-like segregation patterns (CT, TC, T, V-T, and FT). In fact, this index considers the concentration of *jetsam* in the "upper" part of the bed. Nonetheless, in the present case the focus was on the distribution of a particular material of interest, and then, for Top-like segregation patterns, the concentration of this material in the upper 40% of the bed exceeded its total concentration in all the bed, leading to M values greater than unity.

On the other hand, the deviations of "s" index from its limit values can be explained considering its definition, similarly as it was done for the theoretical test. Values greater than 1.00 obey mainly to a considerable decrease of the denominator of Equation 3 ( $\langle h_{\text{large}} \rangle$ ) caused by high amount of the material of interest in the bottom layer. This occurs for Bottom-like segregation patterns where the sum of the product  $m_{\text{P}i}h_i$  is low. On the contrary, values of "s" lower than 0 occur mainly for cases with Top-like segregation patterns where  $\langle h_{\text{large}} \rangle$  is high and S in Equation 3 is less than 1. Thus, the term S - 1(Equation 2) gives negative values.

In Figure 4, as a general trend, it can be seen that as the segregation level  $(\aleph_2)$  increases, the values of M as well as "s" separate from their reference value for perfect mixing. Thus, it can be said that all tested segregation indices are able to recognize segregation to some extent. Indeed, inside the interval [0, 1] of the abscissas there seem to be a correlation between  $\aleph_2$  and the other tested indices. Even though, as it was previously mentioned M and "s" also present values outside their defined limits and, as  $\aleph_2$  takes values only between 0 and 1, another branch of the curve is generated like a piecewise function. Moreover, in Table 7 it is possible to note that M and "s" performances are not very satisfactory if their values are analyzed in detail; the difference in M index for EXP01 and EXP02 is about 20% for similar segregation levels ( $\aleph_2$ ), and it exceeds its maximum expected value. Additionally, despite EXP04 and EXP05 segregation is clearly present, "s" index present values around 0 (perfect mixing).

The time evolution of the studied segregation and mixing indices was also evaluated in two different ways: considering three stages of the drying process (fresh, partially, and completely dried product), and by applying different fluidization times. As it can be noted in Figure 5, the  $\aleph_2$  of the TTSIS shows a good performance in representing the time evolution of the segregation level (it decreases with the product density and the increment of the fluidization time). On the opposite, the 's" index exceeds by far its maximum expected value, when fresh product is utilized and Full Top segregation profile is obtained. Although the M index seems to work quite well in that case, it is not capable to distinguish the segregation pattern in the bed.

Furthermore, the TTSIS was tested with several experimental cases from literature (Table 8) founding that the segregation profiles graphically shown by several authors in fluidized beds can be accurately described by the herein presented segregation indices set. From LT01 to LT05 some examples of segregation patterns exhibited by Wu and Baeyens [11] were presented in terms of TTSIS. Results from LT06 to LT10 (Qiaoqun et al. [15] and Olivieri et al. [16]) evidence that the effect of air velocity, pointed out by the authors, is well reproduced by the TTSIS (i.e. reduction of the segregation level as the air velocity is increased).

Therefore, taking into account the results from literature and experimental ones, it can be said that the TTSIS can be applied for characterizing different segregation cases, for considerably low mass fraction of the material of interest (as the presented experimental values) as well as greater values up to 0.333, independently of the utilized apparatus.

#### 5. Conclusions

A new method for evaluating the segregation of two component mixtures was proposed, and its performance was assessed for different possible experimental situations and segregation profiles. Moreover, two indices from the literature were considered and their performance were compared with the new set of indices.

Segregation results of experiments performed with different number of withdrawn layers could not be compared applying neither M nor "s" indices, as they depend on the number of layers. In addition, they do not allow to distinguish different segregation patterns leading to misleading results when the segregation patterns are near to the extreme cases (such as Pure V, Full Central, or Full Bottom).

On the other hand, using TTSIS for segregation analysis it is possible not only to analyse the results of experiments carried out with the same number of vacuumed layers, but also the results of experiments done with different number of layers. Anyway, in order to obtain a good accuracy of the results in a single experiment, it is recommendable to maintain practically the same layer thickness for all layers.

Therefore, TTSIS was found to be superior to previously proposed indices for quantifying the segregation phenomenon and classifying the segregation patterns. Previous indices are typically used in binary mixtures when the values of mass fractions of floatsam or jetsam are significant, and may give non-reliable results in case of mixtures containing a small fraction of coarse particles in a bed of fine particles, like some of those tested in the present work where a variety of segregation patterns has been observed. TTSIS can be applied successfully with a mass fraction of the material of interest from almost zero to 33% and minimizes possible experimental errors or the discrepancies caused by applying different experimental procedures. Furthermore, the TTSIS can be used in two different ways: compact or full. In the former, only the segregation level and pattern are given (e.g. 0.149-TC), while in the latter, the complete set of indices is expressed (e.g.  $[0.240, 0.371, 0.389]_{0.149}$ ).

Another interesting possibility offered by the TTSIS, is that its use might be easily extended to multicomponent mixtures because it evaluates the distribution of a "material of interest" and does not consider in its calculations the other components of the mixture.

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$p_{\mathrm{I}}$	$p_{\mathrm{M}}$	$p_{\rm S}$	$\aleph_2$	Meaning
0.33	0.33	0.33	0.00	Pure Uniform distribution
0.00	0.00	1.00	1.00	Full Top Segregation
0.00	1.00	0.00	1.00	Full Central Segregation
1.00	0.00	0.00	1.00	Full Bottom Segregation
0.50	0.00	0.50	0.50	Pure V-Segregation

Table 1: TTSIS extreme values.

	$D_{I}$	$p_{\mathrm{M}}$	$^{ m Sd}$	and (other conditions)	$S_{eg}$	<i>then</i> , Segregation type is:
if else if	$\begin{array}{c} 0.33\\ 0.33 \pm 0.03 \end{array}$	$0.33 \\ 0.33 \pm 0.03$	$\begin{array}{c} 0.33\\ 0.33 \pm 0.03 \end{array}$	1 1	nn	Pure Uniform Uniform
else if else if	$\geq 0.98$ $0.55 \leq p_1 < 0.98$	1 1	1 1	$p_{ m S} - p_{ m M} < 0.14$	FB B	Full Bottom Bottom
else if else if	1 1	$\geq 0.98$ $0.55 \leq p_{\rm M} < 0.98$	1 1		C FC	Full Central Central
else if else if	1 1	1 1	$\geq 0.98$ $0.55 \leq p_{\rm S} < 0.98$	$-p_{ m M} < 0.14$	T T	Full Top Top
else if else if else if	$\begin{array}{llllllllllllllllllllllllllllllllllll$	1 1 1	$\begin{array}{l} 0.47 \leq p_{\mathrm{S}} < 0.54 \\ > p_{\mathrm{M}} \ \mathrm{and} > p_{\mathrm{I}} \\ > p_{\mathrm{M}} \end{array}$	1 1 1	V VT VB	Pure V V-Top V-Bottom
else if else if	1 1	$p_{ m I} < p_{ m M} < p_{ m S}$	1 1	1 1	TC BC	Top Central Bottom Central
else if else if	d < 1d < Md	1 1	$p_{ m M}>p_{ m S}>p_{ m I}$ -	1 1	CT CB	Cental Top Central Bottom

Segregation type	$P_{\mathrm{dis},i} =$
Full Bottom	$ \left\{\begin{array}{ll} 1 & i = 1 \\ 0 & i > 1 \end{array}\right. $
Full Top	$\begin{cases} 1 & i = 1 \\ 0 & i > 1 \end{cases}$ $\begin{cases} 1 & i = n_{\rm lyr} \\ 0 & i < n_{\rm lyr} \end{cases}$
Full Control	if $n_{\text{lyr}}$ is even $\begin{cases} 0.5 & i = \frac{n_{\text{lyr}}}{2} \\ 0.5 & i = \frac{n_{\text{lyr}}}{2} + 1 \\ 0 & \forall i \neq \frac{n_{\text{lyr}}}{2} \text{ AND } i \neq \frac{n_{\text{lyr}}}{2} + 1 \end{cases}$
Full Central	if $n_{\text{lyr}}$ is odd $\begin{cases} 1 & i = \left\lceil \frac{n_{\text{lyr}}}{2} \right\rceil \\ 0 & \forall i \neq \left\lceil \frac{n_{\text{lyr}}}{2} \right\rceil \end{cases}$
Pure V	$\begin{cases} 0.5 & i = 1 \\ 0.5 & i = n_{\rm lyr} \\ 0 & 1 < i < n_{\rm lyr} \end{cases}$
Uniform	$\left\{ \begin{array}{cc} rac{1}{n_{ m lyr}} & orall i \end{array}  ight.$

Table 3: Imposed values of the elements of the product mass distribution vector fixed for the segregation pattern studied.

Modified layer	n-th component	Value
Тор	$n_{ m lyr}$	1.4
Top	$n_{ m lyr}$	0.6
Central	$\left\lceil \frac{n_{\text{lyr}}}{2} \right\rceil$	1.4
Central	$\left\lceil \frac{n_{\rm lyr}^2}{2} \right\rceil$	0.6
Bottom	ĩ	1.4
Bottom	1	0.6

Table 4: Variations applied to layer thickness and values given to the different elements of the primary thickness distribution vector.

Table 5: Utilized materials in experimental binary mixtures (data source [14]).

Material	$\rho~(\rm kg/m^3$ )	$d_{\rm SV}~({\rm mm})$	$w_{\rm PT}$ (range)
fresh peas	1088	8.8	0.0476
part.lyo. peas	401	8.8	0.0039 - 0.0138
lyophilized peas <sup>*</sup>	202 / 237	8.8	0.0024 - 0.0109
fresh carrot discs	1050	11.7	0.0476
lyo. carrot discs <sup>*</sup>	112 / 158	10.2 / 9.5	0.0013 - 0.0069
lyo. potato slabs $^*$	176 / 198	8.3 / 12.0	0.0016-0.0046
non-food wheat bran	1469	$6.27{ imes}10^{-1}$ **	-

\*Lyophilized materials with different residual porosity.

\*\*Equivalent diameter at minimum fluidization velocity.

Segr. type	<i>n</i> ,	k	Varied layer	M	"s" index		ΤT	SIS	
imposed	$n_{\rm lyr}$	$k_{\rm co,max}$	and $\%$	IVI	s maex	$p_{\mathrm{I}}$	$p_{\mathrm{M}}$	$p_{\rm S}$	$\aleph_2$
Full Top	2	1.000	_	2.00	-0.333	0.000	0.330	0.670	0.670
Full Top	6	1.000	_	2.50	-0.454	0.000	0.000	1.000	1.000
Full Top	10	1.000	—	2.50	-0.473	0.000	0.000	1.000	1.000
Full Top	100	1.000	_	2.50	-0.497	0.000	0.000	1.000	1.000
Full Bottom	2	1.000	_	0.00	1.000	0.670	0.330	0.000	0.670
Full Bottom	6	1.000	—	0.00	5.000	1.000	0.000	0.000	1.000
Full Bottom	10	1.000	_	0.00	9.000	1.000	0.000	0.000	1.000
Full Bottom	100	1.000	_	0.00	99.000	1.000	0.000	0.000	1.000
Full Central	6	1.000	_	0.50	0.000	0.000	1.000	0.000	1.000
Full Central	10	1.000	—	0.00	0.000	0.000	1.000	0.000	1.000
Full Central	100	1.000	_	0.00	0.000	0.000	1.000	0.000	1.000
Pure V	6	1.000	_	1.25	0.000	0.500	0.000	0.500	0.500
Pure V	10	1.000	—	1.25	0.000	0.500	0.000	0.500	0.500
Pure V	100	1.000	_	1.25	0.000	0.500	0.000	0.500	0.500
Pure Uniform	2	1.000	_	1.00	0.000	0.330	0.330	0.330	0.000
Pure Uniform	6	1.000	_	1.00	0.000	0.330	0.330	0.330	0.000
Pure Uniform	10	1.000	—	1.00	0.000	0.330	0.330	0.330	0.000
Pure Uniform	100	1.000		1.00	0.000	0.330	0.330	0.330	0.000
Full Top	6	1.025	_	2.50	-0.453	0.000	0.000	1.000	1.000
Full Top	10	1.025	_	2.51	-0.473	0.000	0.000	1.000	1.000
Full Top	100	1.025	_	2.51	-0.497	0.000	0.000	1.000	1.000
Pure Uniform	10	1.050	_	1.01	0.002	0.330	0.330	0.330	0.000
Pure Uniform	100	1.050	_	1.01	0.000	0.330	0.330	0.330	0.000
Full Central	6	1.000	Top $+40$	0.20	0.067	0.070	0.930	0.000	0.930
Full Central	6	1.000	Central $+40$	0.70	-0.030	0.050	0.890	0.070	0.840
Full Central	6	1.000	Bottom $+40$	0.70	-0.058	0.000	0.930	0.070	0.930
Full Central	6	1.025	Central -40	0.30	0.039	0.000	1.000	0.000	1.000
Full Central	6	1.025	Bottom $-40$	0.30	0.077	0.130	0.870	0.000	0.870

Table 6: Most representative results of the mathematical test of M, "s" index, and TTSIS.

Case	Vegetable			. <i>M</i>	s			
	, egetable	$p_{\mathrm{I}}$	$p_{\mathrm{M}}$	$p_{\rm S}$	$\aleph_2$	Pattern		
EXP01	lyophilized peas	0.240	0.371	0.389	0.149	TC	1.21	-0.114
EXP02	lyophilized peas	0.274	0.436	0.291	0.162	$\operatorname{CT}$	1.01	-0.010
EXP03	lyo. carrot discs	0.313	0.340	0.346	0.033	U	1.09	0.017
EXP04	lyo. potato slabs	0.335	0.186	0.479	0.293	VT	1.32	-0.099
EXP05	lyo. carrot discs	0.299	0.475	0.226	0.249	CB	0.90	0.058
EXP06	lyo. potato slabs	0.899	0.101	0.000	0.899	В	0.02	3.185

Table 7: TTSIS, M and "s" indices evaluated for the experimental cases (binary mixtures of non-food wheat bran and lyophilized vegetables) in the L35b bed. (Data from [14]).

Case		Source	Materials	q, $w_{qT}$	TTSIS				
Cabo	Ref.	Figure	11100011015	¶, ∞q1	$p_{\mathrm{I}}$	$p_{\rm M}$	$p_{\rm S}$	$\aleph_2$	Pattern
LT01		1a	Larger $(L)/$	S, 0.02	0.030	0.055	0.915	0.885	Т
LT02	[11]	1b	Smaller (S)	S, 0.02	0.048	0.166	0.786	0.737	Т
LT03		1c	(not specified)	S, 0.02	0.111	0.346	$0,\!544$	0.433	TC
LT04		2a	(not specified)	L, 0.024	0.876	0.062	0.062	0.814	В
LT05		2e		L, 0.024	0.377	0.316	0.307	0.070	BC
LT06	[15]	5, 0.58  m/s	Rice husk $(R)/$	R, 0.0582	0.216	0.329	0.455	0.239	$\mathrm{TC}$
LT07	[10]	$5,0.79~\mathrm{m/s}$	Sand $(S)$	R, 0.0582	0.295	0.319	0.387	0.092	TC
LT08		6, 3.2  cm/s	Silica sand (SS)/	SG, 0.2	0.992	0.008	0.000	0.992	$\operatorname{FB}$
LT09	[16]	6, 6.4  cm/s		SG, 0.2	0.637	0.212	0.15	0.487	В
LT10		$6,15.2~\mathrm{cm/s}$	Silica gel (SG)	SG, $0.2$	0.369	0.325	0.306	0.063	BC
LT11	[10]	4	Copper shot $(C)/$	C, 0.2	0.355	0.323	0.323	0.032	U
			Steel shot (S)						

Table 8: TTSIS calculated for data from literature.

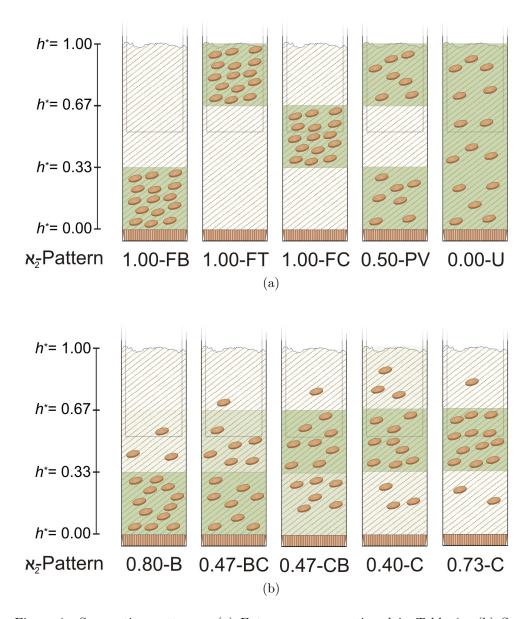


Figure 1: Segregation patterns. (a) Extreme cases mentioned in Table 1. (b) Some intermediate situations. Note that the second and third case present the same value of  $\aleph_2$  but different segregation type; while the fourth and fifth ones present the same segregation type but different  $\aleph_2$ .

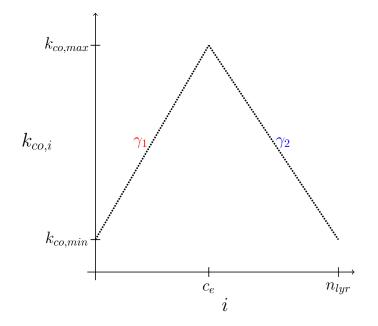


Figure 2: Graphical representation of the variation factor for each layer  $(k_{co,i})$  versus layer number (i), that is, a piecewise defined function composed of two straight lines with positive and negative slopes, with a maximum value,  $k_{co,max}$ , and minimum value,  $k_{co,min}$ .

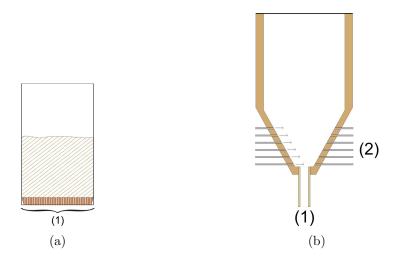


Figure 3: Utilized beds. (a) L35b (350 mm sided fluidized bed) (1) Air distributor. (b) L20spjet (200x100 mm spout-fluid bed). (1) Main air injector, (2) Lateral air injectors.

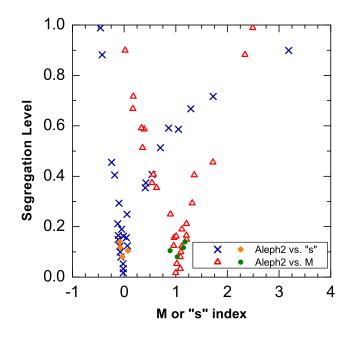


Figure 4: Comparison of the segregation level estimated by  $\aleph_2$  and the two previously proposed indices, for a set of experimental data: M (triangles and circles) and "s" (crosses and diamonds). Data corresponding to experiments carried out using all materials reported in Table 5, in both L35b (blue and red) and L20spjet (orange and green) beds. Data source [14].

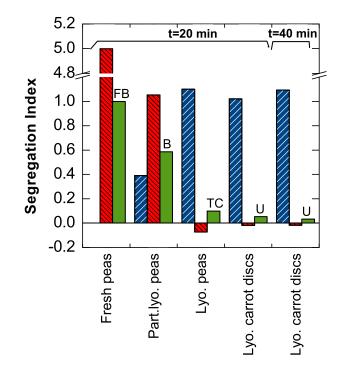


Figure 5: Time evolution of the three considered indices at different level of drying and different fluidization time (red and black slashed, "s"; blue and white slashed, M; green flat,  $\aleph_2$ ). Experiments carried out in the L35b bed, at  $u_{sup,air} = 0.44$  m/s. Data source [14].

### Appendix A. Details for calculating the interpolation function $F_q$ in TTSIS

In the definition of each Third Indicator of the TTSIS, it is involved the accumulated mass of material of interest "q"  $F_q(h^*)$  as a function of  $h^*$ . This parameter is a piecewise function estimating the value of the accumulated mass of material of interest "q" at a given  $h^*$  by linear interpolation. Mathematically it is obtained through the following procedure:

Let the discrete functions  $m_{\text{acq},i}$  and  $h_i^*$ , respectively, the accumulated mass of material of interest "q" from the bed bottom to the layer "i" and the dimensionless height from the bed bottom to the upper limit of layer "i" (Figure A.1). That is,

$$m_{\mathrm{acq},i} = \sum_{s=0}^{i} m_{\mathrm{q},s} \tag{A.1}$$

and

$$h_i^* = \sum_{s=0}^i \Delta h_s^* \tag{A.2}$$

where  $m_{q,s}$  is the mass of material of interest in the layer "s", and  $\Delta h_s^*$  is the dimensionless layer thickness calculated as

$$\Delta h_s^* = \frac{\Delta h_s}{h_{\rm bed}} \tag{A.3}$$

On the other hand, as there is no layer "0" but the first vacuumed layer is denoted with i=1, an index i=0 was defined in order to be coherent with the hereafter definitions. Thus,  $m_{q,0}$  and  $h_0^*$  in Equations A.1 and A.2 are set to 0.

Therefore,  $F_q(h^*)$  is defined as:

$$F_{q}(h^{*}) = \begin{cases} \lambda_{1}(h^{*} - h_{0}^{*}) + m_{acq,0} & 0 \leq h^{*} \leq h_{1}^{*} \\ \vdots \\ \lambda_{i}(h^{*} - h_{i-1}^{*}) + m_{acq,i-1} & h_{i-1}^{*} < h^{*} \leq h_{i}^{*} \\ \vdots \\ \lambda_{n_{lyr}}(h^{*} - h_{n_{lyr}-1}^{*}) + m_{acq,n_{lyr}-1} & h_{n_{lyr}-1}^{*} < h^{*} \leq h_{n_{lyr}}^{*} \end{cases}$$
(A.4)

where  $\lambda_i$  is the slope of the interpolation curve calculated as

$$\lambda_i = \frac{m_{\text{acq},1} - m_{\text{acq},i-1}}{h_i^* - h_{i-1}^*} \tag{A.5}$$

In a more compact form  $F_{\mathbf{q}}(h^*)$  may be written as:

$$F_{q}(h^{*}) = \lambda_{i}(h^{*} - h^{*}_{i-1}) + m_{acq,i-1} \qquad i : h^{*}_{i-1} < h^{*} \leqslant h^{*}_{i} \qquad (A.6)$$

Figure A.2 shows a graphical representation of  $m_{\text{acq},i}$  and  $F_{\text{q}}(h^*)$ .

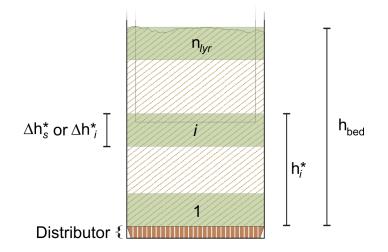


Figure A.1: Bed height, layer height and bed thickness in the settled bed.

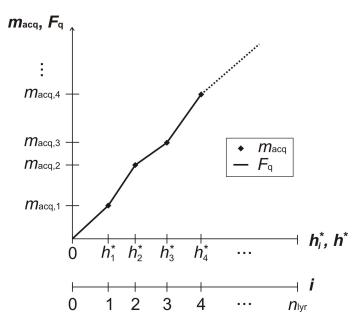


Figure A.2: Schematic representation of the accumulated mass of material of interest  $(m_{\text{acq},i}, \text{discrete function})$  and the interpolation function  $(F_{\text{q}}, \text{continuous})$ .

### Appendix B. Details for extracting information from literature for applying in the TTSIS calculations

In literature the segregation profiles are generally given by means figures in terms of mass fraction of a component of interest (called *jetsam* or *flotsam*) vs. bed height (or vice-versa). Thus, assuming a value for the total mass of material in the bed  $(m_{\rm T})$  and uniform solids bulk density distribution along all the bed, the mass of each component for all the bed layers can be estimated. As for calculating the TTSIS values only mass fractions with respect the total amount of a component of interest are needed, the assumption of  $m_{\rm T}$  does not represent a problem.

Therefore, considering

$$\alpha_{\rm sT}\rho_{\rm sT} = \alpha_{\rm si}\rho_{\rm si} \Rightarrow \frac{\Delta h_i}{h_{\rm bed}} = \frac{m_{\rm si}}{m_{\rm sT}} \tag{B.1}$$

the following equations can be solved:

$$m_{\rm si} = \frac{\Delta h_i}{h_{\rm bed}} m_{\rm sT} \tag{B.2}$$

$$m_{\mathbf{q}i} = w_{\mathbf{q}i}m_{\mathbf{s}i} \tag{B.3}$$

$$m_{\rm Bi} = (1 - w_{\rm qi})m_{\rm si}$$
 (B.4)

$$m_{\rm qT} = \sum_{i} m_{\rm qi} \tag{B.5}$$

$$m_{\rm BT} = \sum_{i} m_{\rm Bi} \tag{B.6}$$

Therefore, for verification the following equation must be matched:

$$w_{\rm AT} = \frac{m_{\rm qT}}{m_{\rm sT}} \approx w_{\rm qT, liter}$$
 (B.7)

Differences between the overall mass fraction calculated and the value given by the source article up to 10 % were accepted.