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## Uncertainty analysis of gamma-ray densitometry applied for gas flow modulation technique in bubble columns

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1	Measuring the gas phase dispersion coefficient in
2	bubble columns with the gas flow modulation
3	technique and gamma-ray densitometry: An
4	uncertainty analysis
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14	Abstract
15	The gas flow modulation technique is a recently proposed approach for measuring the axial gas
16	dispersion coefficient in bubble columns and potentially in other gas-liquid contactors. This study
17	presents a quantitative analysis of the experimental uncertainty that is associated with gamma-
18	ray densitometry and ensemble-averaging of the data. The considered uncertainty sources are the

propagation due to the spatial extent of the detector, and a potential mismatch between modulation and sampling frequencies. The analysis is based on a numerical gamma-radiation propagation model and a Monte Carlo approach to account for statistical uncertainty. The proposed algorithm supports the selection of an optimal total scanning time based on detector size, modulation parameters, involved fluids as well as column and source parameters. The analysis reveals that a mismatch between the modulation and sampling frequencies is most critical while the impact of the other considered uncertainty sources is rather marginal.

statistics of the photon counting process, a mismatch between the modelled and the real radiation

27

### 28 Keywords

29 Gas flow modulation technique, axial dispersion coefficient, gamma-ray densitometry,

30 uncertainty analysis

31

### 32 1. Introduction

Hydrodynamics and mass transfer in gas-liquid contactors are most commonly modelled with the
 one-dimensional axial dispersion model (ADM). It describes the spatiotemporal concentration of
 a species in the liquid or gas phase via the convection-diffusion equation

$$\frac{\partial c}{\partial t} = D \frac{\partial^2 c}{\partial z^2} - u \frac{\partial c}{\partial z}.$$
 1

36 Here, *c* is the concentration of a species in the considered phase, *u* is the superficial velocity of the 37 considered phase and D is the axial dispersion coefficient. A reliable quantification of the axial liquid and gas dispersion coefficients in gas-liquid contactors is crucial for their design and 38 performance assessment. Recently, Hampel [1] proposed the gas flow modulation (GFM) 39 40 technique for the non-invasive measurement of the axial gas dispersion coefficient in bubble columns, which is potentially applicable to other gas-liquid contactors as well. Contrary to the 41 traditional approaches based on tracer substances (e.g., [2, 3]), the GFM technique uses a marginal 42 43 sinusoidal disturbance superimposed on the gas inlet flow rate as virtual tracer. This disturbance 44 introduces a sinusoidal variation of the gas holdup in time, further on referred to as gas holdup 45 wave. Due to the gas dispersion, the gas holdup wave is damped in amplitude and shifted in phase along the column. Mathematically, this can be described in the following way. Assume that the gas 46 flow at the inlet has a constant flow rate plus a marginal sinusoidal change in time. This will 47 produce a modulated gas holdup in the column. Now we consider only the sinusoidal part  $\epsilon'$  of the 48 49 gas holdup. At the inlet we may assume it to be of unit strength and zero phase shift, that is  $\epsilon'(z=0,t) = \cos(\omega t)$ . Further up the column at a position z > 0 we find  $\epsilon'(z > 0,t) =$ 50

51 V(z) cos(ωt + Δφ(z)). That is, the gas holdup wave is damped in amplitude (V) and shifted in
52 phase (Δφ).

53 The one-dimensional axial dispersion model for the gas holdup is given in the time domain as

$$\frac{\partial \epsilon(z,t)}{\partial t} = D_{\rm G} \frac{\partial^2 \epsilon(z,t)}{\partial z^2} - u_{\rm G}^* \frac{\partial \epsilon(z,t)}{\partial z}, \qquad 2$$

54 and in the frequency domain as

$$j\omega E(z,\omega) = D_{\rm G} \frac{\partial^2 E(z,\omega)}{\partial z^2} - u_{\rm G}^* \frac{\partial E(z,\omega)}{\partial z},$$
3

where z is the axial distance from the gas sparger,  $D_{\rm G}$  is the axial gas dispersion coefficient,  $u_{\rm G}^*$  is the average bubble rise velocity,  $\omega = 2\pi f$  is the modulation frequency and *E* is the time-domain Fourier transform of the axial gas holdup distribution. Solving Equation 3 analytically one can derive the following expressions for *V* and  $\Delta \phi$  between two axial planes with distance  $\Delta z$  [1]:

$$V = \exp\left(\frac{u_{\rm G}^*}{2D_{\rm G}}\left[1 - \frac{1}{\sqrt{2}}\sqrt{1 + \sqrt{1 + \frac{16\omega^2 D_{\rm G}^2}{u_{\rm G}^{*\,4}}}}\right]\Delta z\right),$$
4

$$\Delta \phi = -\frac{u_{\rm G}^*}{D_{\rm G}\sqrt{8}} \left[ \sqrt{\left[ \sqrt{1 + \frac{16\omega^2 D_{\rm G}^2}{{u_{\rm G}^*}^4}} \right] - 1} \right] \Delta z.$$
 5

Both equations are needed for determining the axial gas dispersion coefficient. In fact, the limited sensitivity of Equation 5 to a change in  $D_G$  does not allow reliable calculation of  $D_G$ , as shown by Marchini et al. [4]. In addition, multiple solutions to Equation 4 in terms of  $D_G$  exist in at least part of the domain. In order to use Equations 4 and 5 for calculating  $D_G$  the values of the amplitude and phase of the gas holdup wave must be measured at least at two axial positions in the column. This was first demonstrated by Döß et al. [5] using gamma-ray densitometry as illustrated in the simplified scheme in **Figure 1a**.

Recently, Marchini et al. [4] introduced a theoretical analysis and quantification of the inherentuncertainty caused by the use of the axial dispersion model for describing the gas holdup wave.

However, this comprehensive analysis did not take into account the contribution of the
experimental uncertainty to the measured amplitude attenuation and phase shift. Thus, the
objective of this paper is to fill this gap.



71

Figure 1. Simplified scheme of the experimental setup needed for axial gas dispersion
 coefficient measurement using gamma-ray densitometry with (a) one or (b) two
 synchronized detector elements and radiative sources.

An alternative arrangement to the one represented in **Figure 1a** is composed of two radiative sources and two detector elements. The detectors sample simultaneously at two different axial positions of the column and are synchronized with the same clock (Clock 2). The gas flow modulator, instead, relies on another clock (Clock 1). Both clocks are again synchronized with each other. This configuration allows measuring at the same time at both axial positions, reducing the impact of possible deviations in the process and the time needed to perform the measurements. A representative scheme is provided in **Figure 1b**.

In both configurations, the parameters contributing to experimental uncertainty are manifold and dependent on the specific setup. This work considers uncertainty sources that are common to most gamma-ray densitometry setups, i.e. statistics of the photon counting process, a mismatch between the modelled and the real radiation propagation due to the spatial extent of the detector,
and a potential mismatch between modulation and sampling frequencies. In this study, numerical
experiments are preferred over real ones due to the high number of repetitions needed for each
experiment. Furthermore, such a numerical approach enables to decouple the above-mentioned
uncertainty sources that act together in real experiments.

90 The proposed approach is used to simulate the gamma-photons registered by a gamma-ray 91 detector to quantify the impact of the selected uncertainty sources depending on the operating 92 conditions as well as on column characteristics. The algorithm also supports optimizing the 93 scanning time and sampling frequency for cost-efficient experiments.

94

### 95 2. Ensemble-averaging gamma-ray densitometry and uncertainty sources

Gamma-ray densitometry is based on the measurement of the linear attenuation of radiation within an object. Therefore, an isotopic source and a photon-counting detector are arranged opposite to each other at the object of interest such that radiation from the source passes the object and is registered by the detector. The detector counts single photons arriving from the source. Hence, in the following we denote by *N* the number of photons being registered in a given time interval  $\Delta T$ . In case of a gas-liquid contactor, we find the following relationship for radiation attenuation:

$$\langle N_{\rm GL} \rangle = \langle N_{\rm G} \rangle \exp(-\mu_{\rm L} l(1-\epsilon)).$$
 6

Here,  $\langle N_{GL} \rangle$  is the expected number of photons registered per time interval for an operating contactor (note, that subscript *G* stands for gas and *L* stands for liquid),  $\langle N_G \rangle$  is the expected number of photons one would register per time interval for the contactor containing no liquid. Furthermore,  $\mu_L$  is the linear attenuation coefficient of the liquid, *l* is the total length of the process space the radiation beam passes and  $\epsilon$  is the gas holdup within the beam path. Here, two technical remarks are necessary. First, the derivation of Equation 6 assumes that the linear attenuation 109 coefficient of the gas phase  $\mu_{\rm G} = 0$ . For contactors operating at very high pressures we may have 110  $\mu_{\rm G} > 0$ , but this can be considered in a straightforward way. Second, in gamma-ray densitometry 111 the detector will inevitably record some additional natural radiation in addition to the radiation 112 from the isotopic source. It is therefore assumed that this constant offset is known and corrected. 113 Every radiation-emission-detection process is associated with a statistical uncertainty. The count 114 number in a time interval fluctuates and the statistical distribution of the count number is 115 described by a Poisson distribution

$$P(N) = \frac{\langle N \rangle^N}{N!} \exp(-\langle N \rangle), \qquad 7$$

116 where *P*(*N*) is the probability of detecting *N* photons if  $\langle N \rangle$  are expected. Note, that *N* ∈ N and 117  $\langle N \rangle \in \mathbb{R}$ .

To reduce the impact of the statistics of the photon counting process on measured amplitude and phase, Döß et al. [5] applied a lock-in detection scheme, that synchronises the detector data acquisition with the gas flow modulation. The detection events were then ensemble-averaged by counting them within  $n_s = f_s/f$  equidistant scanning intervals, where  $f_s$  and f are the sampling and modulation frequencies, respectively. **Figure 2** illustrates this schematically. From here on, ensemble-averaged data are indicated with a tilde.



Figure 2. Schematic of the ensemble-averaging procedure, in which (a) the original detector
 data stream is subdivided into single modulation periods that are (b) subsequently averaged to
 reduce statistical noise. This way, the modified modulation wave is obtained.

As a result, one obtains the ensemble-averaged count number distributions for one modulation period containing  $n_s$  values. This distribution is logarithmized to obtain gas holdup data. A cosine function is then fitted to the resulting data, whose amplitude and phase are determined by a leastsquares criterion. Note that if there was no logarithmization one could also do a Fourier analysis to obtain amplitude and phase.

133 Using gamma-ray densitometry one needs to consider its specific sources of uncertainty. First, the 134 gamma-ray detector is commonly assumed to be point-like. However, in reality, the extent of the 135 detector is a source of uncertainty. In case of a circular object (i.e. a column), the penetration length changes along the detector width while the gas holdup wave changes its amplitude and 136 137 phase along the detector height. Another uncertainty source addressed in this study is a mismatch between the modulation and sampling frequencies and vice versa. This may be due to 138 139 insufficiently synchronized clocks. Any mismatch between the modulation and sampling frequencies reduces the reliability of the reconstructed amplitude and phase. 140

141

### 142 **3. Methodology**

In this section, we present an approach that predicts the gamma-photon rate registered by the detector at a certain axial position. At a fixed axial position, the gas holdup for the sinusoidal modulation is a function of time, that is

$$\epsilon(t) = \bar{\epsilon} + A_{\epsilon} \cos(\omega t + \phi), \qquad 8$$

146 where  $\bar{\epsilon}$  is the time-averaged gas holdup,  $A_{\epsilon}$  is the modulation amplitude, and  $\phi$  is the modulation 147 phase. Substituting Equation 8 in Equation 6, the expected number of counts in time for given 148 amplitude and phase of the gas holdup wave is

$$\langle N_{\rm GL}(t) \rangle = \langle N_{\rm G} \rangle \exp(-\mu_{\rm L}(1 - \bar{\epsilon} - A_{\epsilon} \cos(\omega t + \phi))l) \,.$$

Equation 9 is used to model gamma-ray densitometry for assumed modulation parameters ( $\bar{\epsilon}, A_{\epsilon}$ ,  $\phi, \omega$ ), initial count number ( $N_{\rm G}$ ), column geometry (l) and working fluids ( $\mu_{\rm L}$ ). Count numbers are simulated for  $n_{\rm tot}$  equal-size time intervals between t = 0 and  $t = t_{\rm tot}$ . For each time interval we compute a count number realization  $N_{\rm GL}$  by drawing a Poisson-distributed number (Equation 7) using the expectation value computed from Equation 9. Having done so for all  $n_{\rm tot}$  sampling points we perform the ensemble averaging and logarithmization which gives the gas holdup distribution for one ensemble-averaged period as

$$\tilde{\epsilon}_{i}^{*} = \frac{1}{\mu_{\rm L} l} \log \left( \frac{\widetilde{N}_{\rm GL,i}}{\langle N_{\rm G} \rangle} \right) + 1.$$
 10

156 Eventually, these data are then fitted by a cosine function as

$$\epsilon^*(t) = \bar{\epsilon}^* + A_{\epsilon}^* \cos(\omega t + \phi^*), \qquad 11$$

obtaining the amplitude  $A_{\epsilon}^*$  and phase  $\phi^*$ . The comparison between  $A_{\epsilon}^*$  and  $A_{\epsilon}$  and between  $\phi^*$  and  $\phi$  provides a quantification of the uncertainty caused by the statistics of the photon counting process on amplitude damping and phase shift, respectively.

160 Since the fitting of the cosine function is only based on the specific values of  $\tilde{N}_{GL}$ , several 161 repetitions of the same experiment are necessary to obtain a reliable assessment, as typical for 162 Monte Carlo approaches. The results obtained this way offer more than 95% confidence level. It 163 should be noted that the deviation caused by statistics of the photon counting process is physically 164 inherent to all configurations and setups.

165 The detector size plays a role for the statistics of the photon counting process and further the 166 spatial extent of the detector may have the consequence that radiation attenuation does no longer 167 fit to a simple one-beam model. The count rate increases with the extension of the detector. 168 Considering an empty process space, the number of detection events in a time interval  $\Delta T$  is given 169 by

$$\langle N_{\rm G} \rangle = \frac{\Phi S \Delta T}{4\pi L^2},$$

170 where  $\Phi$  is the gamma-radiation flux of the source (photon emission rate into the full solid angle 171 of  $4\pi$  sr) and *S* is the detector area. The geometrical parameters are illustrated in **Figure 3**. A 172 bigger detector improves the statistics (since it increases  $\langle N_G \rangle$ ). However, an increased detector 173 size results in changes in the real object length accompanied by changes in the gamma-ray 174 attenuation along the horizontal direction (detector width  $L_{d,y}$ ). Additionally, the detector height 175  $L_{d,z}$  affects the measurement since the amplitude and phase of the gas holdup wave change along 176 the vertical axis of the contactor.



177

Figure 3. Symbols used in Section 5 (parameters used describing a single detector element is
labelled in red) and illustration of the effect of the detector height (a) and width (b).

180

### 181 **4. Statistical uncertainty in the photon counting process**

In this section, we assess the uncertainty in amplitude and phase caused by the statistics of the photon counting process. For this purpose, the detector is assumed small enough so that the penetration length is constant and equal to the length of the beam in the process space. Here, amplitude and phase do not change within the axial limits of the detector. The effect of the detector size will be analysed in Section 5. Considering a measurement time  $t_{tot}$  and a sampling frequency  $f_s$ , the total expected number of counts for each interval  $t_i$  of the modulation period (i.e., here  $0 \le$  $t_i \le 1/f$ ) is (see Equations 9 and 12)

$$\langle N_{\rm GL,i} \rangle = \frac{t_{\rm tot} f}{f_{\rm s}} \langle N_{\rm G} \rangle \exp(-\mu_{\rm L} l(1-\epsilon_{\rm i})).$$
 13

189 Defining

$$K_{1} = \frac{t_{\text{tot}}f}{f_{\text{s}}} \langle N_{\text{G}} \rangle \exp\left(K_{2}\left(1 - \frac{1}{\bar{\epsilon}}\right)\right), \qquad 14$$

$$K_2 = \mu_{\rm L} \bar{\epsilon} l \,, \tag{15}$$

190 Equation 13 can be rewritten as

$$\langle N_{\rm GL,i} \rangle = K_1 \exp(K_2 A \cos(\omega t_i + \phi)), \qquad 16$$

191 where  $K_1$  and  $K_2$  are independent of the considered time interval. The above-presented algorithm 192 is used to evaluate the effect of  $K_1$  and  $K_2$  on the standard deviations in amplitude damping and 193 phase-shift, which are reported in **Figure 4** and defined as

Dev
$$A = \frac{1}{A} \sqrt{\frac{1}{e} \sum_{i=1}^{e} (A_i^* - A)^2},$$
 17

Dev
$$\phi = \frac{1}{\phi} \sqrt{\frac{1}{e} \sum_{i=1}^{e} (\phi_i^* - \phi)^2},$$
 18

194 where *A* and  $\phi$  are the expected values of amplitude and phase, while  $A_i^*$  and  $\phi_i^*$  are amplitude 195 and phase obtained from each numerical experiment. The number of repetition for each 196 experiment is indicated as *e*.

**Table 1** gives values of  $K_1$  and  $K_2$  for typical configurations and working fluids. An analysis of the effect of *A* and  $\phi$ , for constant values of  $K_1$  and  $K_2$  is reported in the **Supplementary Material S1**. This analysis showed that the phase of the gas holdup has only a limited effect on the uncertainty on the phase itself. On the contrary, a higher amplitude substantially reduces the uncertainty of both in terms of amplitude and phase. However, as discussed by Marchini et al. [4], the initial 202 modulation amplitude should be kept below 15% to avoid an effect of the modulation on the203 hydrodynamics of the column.

204

	Case N.	$\mu_{\rm L} l$	$\langle N_{\rm G} \rangle$	$t_{\rm tot} f f_{\rm s}^{-1}$	$ar{m{\epsilon}}$	K <sub>1</sub>	K <sub>2</sub>		
	(-)	(-)	(cps)	(s)	(-)	(s <sup>-1</sup> )	(-)		
	1		$4.9 \cdot 10^{3}$			783			
	2		9.8 · 10 <sup>3</sup>			1566			
	3 4	F	$2.7 \cdot 10^{4}$	14.4	0.1	4349	05		
		5	6.1 · 10 <sup>4</sup>		14.4 0.1	0.1	9785	0.5	
	5		1.3 · 10 <sup>5</sup>				19569		
	6		1.5 · 10 <sup>5</sup>			24462			
	7		1.3 · 10 <sup>3</sup>	17	17	1 7		941	
	8	1		1.7		1882			
	9		$2.5 \cdot 10^{3}$	3.5	0 1 5	3764	0.15		
	10			7.0	0.15	7529	0.15		
	11		4.0 1.03	7.0		15058			
	12		4.9 · 10°	14.4		30117			
	13		1.3 · 10 <sup>3</sup>	17		895			
	14			1.7		1790			
	15	15 1 16 1 17 18	2.5 · 10 <sup>3</sup>	3.5	0.1	3581	0.10		
	16			7.0	0.1	7162	0.10		
	17		4.0 1.03	7.0		14324			
	18		$4.9 \cdot 10^{3}$	14.4		28648			
			1	1			1		

**Table 1.** Examples of obtained  $K_1$  and  $K_2$  for different configurations.



Figure 4. Deviation of amplitude and phase as a function of  $K_1$  and for different values of  $K_2$ ( $A = 0.15, \phi = 0.5$ ).

Figure 4 shows a decrease in both deviations increasing  $K_1$  and  $K_2$ . However, it should be noted that  $K_1$  itself is dependent on  $K_2$  and it decreases significantly when the latter is increased.

210

### 211 5. Effect of the spatial extent of the detector

As pointed out in Section 4, the detector size influences the value of  $K_1$ . In particular, a larger detector can be used to increase the photon count rate and, therefore, reduce the deviation in amplitude and phase. However, the changes along the detector surface in the length of the radiation beam inside of the process space and in the gas holdup were not considered. In this section, the impact of these assumptions is quantified in terms of additional uncertainty, neglecting the effect of the statistics of the photon counting process.

The problem is approached dividing the detector into a number of small rectangular sections with columns (index j) and rows (index k) and center coordinates  $P_{jk}^{(c)}\left(x_{jk}^{(c)}, y_{jk}^{(c)}, z_{jk}^{(c)}\right)$  as shown in **Figure 3a**. Accordingly, each detector section contributes with

$$N_{\rm GL,jk} = N_{\rm G,jk} \exp\left(-(1-\bar{\epsilon})\mu_{\rm L}L_{\rm jk}\right) \exp\left(A_{\epsilon,0}\mu_{\rm L}\int_{0}^{l_{\rm jk}}V(l')\cos\left(\omega t + \Delta\phi(l')\right)dl'\right).$$
19

If the ray penetrates and leaves the column (considered as an empty cylindrical process space) in  $P_{jk}^{(1)}(x_{jk}^{(1)}, y_{jk}^{(1)}, z_{jk}^{(1)})$  and  $P_{jk}^{(2)}(x_{ji}^{(2)}, y_{ji}^{(2)}, z_{ji}^{(2)})$ , respectively, the penetration length is

$$l_{jk} = \sqrt{\left(x_{jk}^{(2)} - x_{jk}^{(1)}\right)^2 + \left(y_{jk}^{(2)} - y_{jk}^{(1)}\right)^2 + \left(z_{jk}^{(2)} - z_{jk}^{(1)}\right)^2}.$$
 20

The total number of counts reaching the detector is given by the sum of the counts reaching eachsection, that is

$$N_{\rm GL} = F \sum_{k=1}^{\rm K} \sum_{j=1}^{\rm J} \frac{1}{L_{jk}^2} \exp(-(1-\bar{\epsilon})\mu_{\rm L}l_{jk})$$

$$\cdot \exp\left(A_{\epsilon,0}\mu_{\rm L}C \int_{z_{jk}^{(1)}}^{z_{jk}^{(2)}} \exp(H_1z)\cos(\omega t + H_2z + \phi_0)\,{\rm d}z\right),$$
21

225 where

$$F = \frac{\Phi S}{4\pi f_{\rm s}}$$
 22

$$H_{1} = \frac{u_{\rm G}^{*}}{2D_{\rm G}} \left[ 1 - \frac{1}{\sqrt{2}} \sqrt{1 + \sqrt{1 + \frac{16\omega^{2}D_{\rm G}^{2}}{u_{\rm G}^{*4}}}} \right],$$
 23

$$H_{2} = -\frac{u_{G}^{*}}{D_{G}\sqrt{8}} \left[ \sqrt{\left[ \sqrt{1 + \frac{16\omega^{2}D_{G}^{2}}{u_{G}^{*}}} \right] - 1} \right],$$
 24

$$C = \sqrt{1 + \left(\frac{x_{jk}^{(2)} - x_{jk}^{(1)}}{z_{jk}^{(2)} - z_{jk}^{(1)}}\right)^2 + \left(\frac{y_{jk}^{(2)} - y_{jk}^{(1)}}{z_{jk}^{(2)} - z_{jk}^{(1)}}\right)^2}$$
25

226 and

$$L_{jk} = \sqrt{x_{jk}^{(c)^2} + y_{jk}^{(c)^2} + z_{jk}^{(c)^2}}.$$
26

### 227 The detailed derivation of Equation 21 is given in the **Supplementary Material S2**.

Equation 21 was solved numerically for given values of F/S,  $H_1$ ,  $H_2$ ,  $A_{\epsilon,0}\mu_L$  and for a given relative position between the column and the measurement system. The solution was obtained for different normalized values of the detector width and height ( $L_{d,y}/l$  and  $L_{d,z}/l$  in **Figure 5**) and for each time step  $t_i$  considering a single modulation period. The obtained number of counts was then converted into holdup data as

$$\epsilon_{i}^{*} = 1 + \frac{1}{\mu_{\rm L}l} \ln\left(\frac{L^2 N_{\rm GL,i}}{F}\right) = \bar{\epsilon}^{*} (1 + A^* \cos(\omega t_i + \phi^*))$$
<sup>27</sup>

and fitted by a cosine function. The obtained amplitude, phase and average holdup are then compared with their expected values  $\bar{\epsilon}$ ,  $\phi = H_2 z + \phi_0$  and  $A = A_{\epsilon,0} \exp(H_1 z)$ . Figure 5 reports the obtained results for

$$DevA = \frac{A^* - A}{A},$$
 28

$$Dev\phi = \frac{\phi^* - \phi}{\phi},$$
 29

$$Dev\bar{\epsilon} = \frac{\bar{\epsilon}^* - \bar{\epsilon}}{\bar{\epsilon}}.$$
 30





Figure 5. Deviations on (a-b) amplitude, (c-d) phase and (e-f) average values of the gas holdup wave, as a function of detector width and height normalized by the length of the radiation beam in the process space ( $H_1 = -0.3669 \text{ m}^{-1}$ ,  $H_2 = 1.9502 \text{ m}^{-1}$ ,  $F/S = 8 \cdot 10^5 \text{ m}^{-2}$ ,  $A_{\epsilon,0}\mu_L =$  $0.1 \text{ m}^{-1}$ , L = 0.4 m, a = 0.3 m).

Other examples for different values of  $H_1$  and  $H_2$  are reported in the **Supplementary Material** 241 **S3**. The results show that the detector size has only a marginal influence on amplitude, phase and 242 243 average value of the measured gas holdup wave. Thus, larger detectors (or combining several smaller ones) are recommended to reduce the total measurement time. It should be kept in mind, 244 however, that for large detectors the deviation on the average holdup value might be non-245 246 negligible (see **Figure 5e**,**f**). The average holdup is linked with the bubble swarm velocity via  $u_{\rm G}^*$  =  $u_{\rm G}/\bar{\epsilon}$ , and a deviation on the bubble swarm velocity causes an additional deviation on the obtained 247 248 axial gas dispersion coefficient, which is not considered here. This deviation has been analysed by Marchini et al. [4]. 249

250

### 251 6. Impact of a frequency mismatch on measured amplitude and phase

When quartz-controlled timers are used in the measurement system, no relevant shift is expected between the sampling and modulation frequencies. However, to highlight the importance of correct synchronization the impact of a frequency shift is addressed here.

# 6.1 Quantification of the impact of a frequency mismatch on measured amplitude and phase

The deviation caused by a mismatch between the sampling and modulation frequencies is quantified modifying the algorithm proposed in Section 3. In this case, the real modulation and sampling frequencies  $f^*$  and  $f_s^*$ , respectively, are used to simulate the count data set sampled by the detector. In order to quantify the deviation, this obtained data set is then wrongly ensembleaveraged based on the expected number of intervals per modulation period  $n_s$ . Defining

$$Devn_{s} = \frac{n_{s}^{*} - n_{s}}{n_{s}},$$
31

where  $n_s^* = f_s^*/f^*$ , **Figure 6** reports an example of the impact of this frequency mismatch on the determined amplitude and phase in terms of Dev*A* and Dev $\phi$ , as defined in Equations 28 and 29, respectively. For the sake of simplicity, signals free from other uncertainty sources are used here and  $f_s^*/f^*$  is assumed integer.



Figure 6. Impact of an unexpected deviation in the number of intervals per modulation period on amplitude and phase of the gas holdup wave (for A = 0.15,  $\phi = 1.5$  rad,  $K_1/(t_{tot}f) =$ 19.9 s<sup>-1</sup>,  $K_2 = 0.1$ ). |DevA| and |Dev $\phi$ | generally increase with |Dev $n_s$ |. Points deviating from this trend are highlighted in red.

The impact of the frequency shift on the ensemble-averaged signal increases with the total scanning time and with the shift magnitude, reaching 100%. The few points deviating from the main trend in **Figure 6** (highlighted in red) are due to the periodicity of the cosine function or due to the inadequacy of the fitting function, as amplitudes approach zero. **Figure 6** shows that even a minor undetected change can cause the failure of the ensemble-averaging approach, especially for long total scanning times. An analytic prediction of the deviation caused in such a scenario is provided in the **Supplementary Material S4**.

278

266

### **6.2 Determining the modulation frequency from available detector data**

If the modulation frequency is not known, it can be determined employing a discrete Fouriertransform to the detector data. This, however, introduces an uncertainty in the determined

frequency, which is quantified in this subsection. The maximum uncertainty of the determined
frequency corresponds to the resolution of the frequency vector of the discrete Fourier transform
equal to

$$\Delta f = \frac{1}{t_{\text{tot}}}.$$
32

Based on Equation 31, the resolution of the Fourier transform causes a maximum deviation of  $n_s$ for given sampling frequency and total scanning time given by

$$Devn_{s} = \frac{1}{t_{tot}f_{s}}n_{s}^{*}.$$
33

287 For small deviation values

$$\mathrm{Dev}n_{\mathrm{s}} \cong \frac{1}{t_{\mathrm{tot}}f_{\mathrm{s}}}n_{\mathrm{s}}.$$
 34

Therefore, the deviation caused by the limited resolution of the fast Fourier transform decreases increasing  $t_{tot}f_s$  and increases linearly with  $n_s$ .

290 Figure 7 shows examples of how the uncertainty of the determined frequency (defined as in

Equation 31) changes with the sampling time and the number of intervals per period  $n_s$ .



292

**Figure 7.** Deviation of the measured number of intervals per modulation period as a function of

294

the expected value.

The shortcomings of the discrete fast Fourier transform are well-known in the literature and several techniques have been proposed to reduce the uncertainty of the determined frequency, for example by Kanatov et al. [6] and by Gasior and Gonzalez [7]. More specifically, the Gaussian interpolation of the frequency spectrum, proposed by Gasior and Gonzalez [7], is proved to reduce this uncertainty by up to three orders of magnitude. The application of these techniques allows reducing the uncertainty way more than increasing the total scanning time and should therefore be preferred.

Even a very small uncertainty of 0.1% on the sampling over the modulation frequency ratio can cause the failure of the approach (see **Figure 6**). Thus, care must be taken in selecting proper clocks for measurement system and signal detection. In addition, techniques to reduce the deviation due to the discrete nature of the fast Fourier transform should be implemented in the analysis, if the modulation frequency is not exactly known.

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### 308 7. Uncertainties of the axial dispersion coefficient

309 When performing measurements, the average gas holdup value as well as the amplitude and phase 310 of its wave at certain axial positions are obtained. This study quantifies the experimental 311 uncertainties associated with these parameters, when gamma-ray densitometry is used as 312 measurement technique. However, the experimental uncertainty for amplitude damping and 313 phase-shift obtained from this study must be combined with the inherent uncertainty (related to the assumptions involved in Equations 4 and 5) that has been analyzed recently by Marchini et al. 314 315 [4]. This allows obtaining the resulting uncertainty of the measured axial dispersion coefficient. For clarity, a scheme of contributions of inherent and experimental uncertainty is provided in 316 317 Figure 8.

Gas flow modulation experiments

**Axial Dispersion Model** 



318



Marchini et al. [4] analyzed the derivative of the amplitude damping and phase-shift with respect to the axial dispersion coefficient. These are functions of the modulation and geometric parameters, the bubble rise velocity and the axial gas dispersion coefficient itself. Therefore, the same uncertainty of amplitude damping and phase-shift leads to different uncertainties on the axial gas dispersion coefficient considering different conditions.

As an example, Figure 8 shows the sensitivity of the amplitude damping and phase-shift to theaxial dispersion coefficient for different bubble rise velocities.



**Figure 8.** Sensitivity of (a) the amplitude damping and (b) the phase-shift to the axial gas dispersion coefficient obtained for different bubble rise velocities (f = 0.4 Hz,  $\Delta x = 0.2$  m).

### 331 8. Conclusions

We presented a study on the uncertainty of gamma-ray densitometry applied to the gas flow modulation technique and considered therefore the statistics of the photon counting process. The analyses shows that the measuring uncertainty, in principle, can be reduced by increasing the modulation amplitude, while modulation and sampling frequencies have no effects. However, this practice should be discouraged, since the modulation amplitude should always be kept below 0.15 as shown by Marchini et al. [4].

A reliable and systematic approach was proposed for quantifying the experimental uncertainty on the determined amplitude and phase of the gas holdup wave for various setup characteristics and operating conditions. The uncertainty connected with the detector size was quantified, too. In this way, in addition to increasing the total scanning time, the use of larger detectors (or the averaging of more adjacent detector elements) is a proper mean for reducing the uncertainty caused by the photon counting process. In this case, however, a well-centred alignment between source, column and detector is crucial.

It was further shown that the sampling frequency can be directly determined from the obtained data, applying a discrete fast Fourier transform. However, even an unexpected minimum deviation in the number of intervals per modulation period (i.e. in the frequency) can cause a failure of the ensemble-averaging approach. For this reason, extreme care should be taken in selecting the applied clocks and the limited resolution of the discrete Fourier transform should be kept in mind.

Altogether, the proposed algorithm and derived guidelines represent a powerful support for designing gamma densitometry measurement systems for future applications of the GFM (including characteristics of gamma-ray source and detector, geometry of the apparatus and needed performances of the clocks), as well as for selecting appropriate total scanning time, sampling frequency and modulation frequency.

356

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359

### Nomenclature

а	Distance between source and column along the x axis for y=0	m
	and z=0.	
Α	Amplitude of the sinusoidal gas holdup wave	
$A_{\epsilon}$	Amplitude of the sinusoidal gas wave times average gas holdup.	
$A_{\epsilon,0}$	Initial amplitude of the sinusoidal gas wave times average gas	
	holdup.	
DevA	Deviation of the measured amplitude of the gas holdup wave	
С	Concentration of selected species in the studied phase	mol m <sup>-3</sup>
С	Parameter used in Section 5, defined as $C =$	m
	$\sqrt{1 + \left(\frac{x_{jk}^{(2)} - x_{jk}^{(1)}}{z_{jk}^{(2)} - z_{jk}^{(1)}}\right)^2 + \left(\frac{y_{jk}^{(2)} - y_{jk}^{(1)}}{z_{jk}^{(2)} - z_{jk}^{(1)}}\right)^2}$	
D	Axial gas dispersion coefficient	$m^2s^{-1}$
е	Number of repetitions of a single experiment	
Ε	Time-domain Fourier transform of the gas holdup distribution	
f	Modulation frequency	Hz
$\Delta f$	Resolution of the frequency vector of the Fourier transform	Hz
fs	Sampling frequency	Hz
F	Parameter used in Section 5, $F = \frac{\Phi S}{4\pi f_s}$	
$H_1$	Parameter used in Section 5 defined as	$m^{-1}$

$$H_1 = \frac{u_G^*}{2D_G} \left[ 1 - \frac{1}{\sqrt{2}} \sqrt{1 + \sqrt{1 + \frac{16\omega^2 D_G^2}{u_G^*}}} \right]$$
 $H_2$ Parameter used in Section 5 defined as $m^{-1}$  $H_2 = -\frac{u_G^*}{D_G\sqrt{6}} \left[ \sqrt{\left[ \sqrt{1 + \frac{16\omega^2 D_G^2}{u_G^*}} \right] - 1} \right]$  $K_1$ Parameter used in Section 4 defined as  $K_1 =$ cps $\frac{t_{out}f}{f_s} (N_G) \exp\left(K_2\left(1 - \frac{1}{\epsilon}\right)\right)$  $K_2$ Parameter used in Section 4 defined as  $K_2 = \mu_L \epsilon l$  $l$  $l$ length of the radiation beam in the process spacem $L$ Distance between source and detectorm $L_{dx}$ Detector widthm $L_{dy}$ Detector heightm $n$ Scanning intervalm $n_s$ Number of scanning intervals per modulation periodDevens $N_{tot}$ Total number of intervals in the total measurement time $N$  $N$ Number of counted photonscps $\langle N_G \rangle$ Expected number of counts for the gas-filled columncps $\langle N_G \rangle$ Expected number of counts for the gas-liquid-filled columncps $R_{GL}$ Ensemble-averaged number of counts for the gas-liquid-filled columncps $R_{GL}$ Probability $M$  $M$ 

$P^{(c)}$	Center of the detector element with coordinates $x^{(c)}, y^{(c)}, z^{(c)}$	
<i>P</i> <sup>(1)</sup>	Point where the considered gamma-ray penetrates the column	
	with coordinates $x^{(1)}, y^{(1)}, z^{(1)}$	
<i>P</i> <sup>(2)</sup>	Point where the considered gamma-ray exits the column with	
	coordinates $x^{(2)}, y^{(2)}, z^{(2)}$	
S	Detector surface ( $S = L_{d,z}L_{d,y}$ )	m <sup>2</sup>
t	Time	S
t <sub>tot</sub>	Total measurement time	S
u	Superficial velocity	ms <sup>-1</sup>
$u_{ m G}^{*}$	Bubble rise velocity	ms <sup>-1</sup>
V	Amplitude damping between two axial positions	
<i>x</i> , <i>y</i>	Coordinate axis with origin corresponding to the gamma-source	m
Ζ	Axial coordinate of the bubble column (distance from the	m
	sparger)	
$\Delta z$	Axial distance between the two measurement points	m

### **Greek letters**

μ	Attenuation coefficient	$m^{-1}$
E	Expected gas holdup wave	
ε	Expected ensemble-averaged gas holdup wave	
$\bar{\epsilon}$	Expected gas holdup wave averaged over the entire	
	measurement time	
Devē	Deviation of the measured average gas holdup	
$\phi$	Phase of the gas holdup wave	rad
$\Delta \phi$	Phase-shift in the gas holdup wave between the measurement	rad
	points	
Dev $\phi$	Deviation on the phase of the gas holdup wave	

Φ	Photon intensity	cps sr <sup>1</sup>
ω	Angular modulation frequency	rad s <sup>-1</sup>

### **Subscripts**

i, j, w, k	Indexes
G	Gas phase
L	Liquid phase

### **Superscripts**

j,i	Refers to detector element (j,i)
*	Measured value (in contrast to the expected value)

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