Quantitative Flash X-ray Imaging of Liquid Mass Distribution in Optically Dense Sprays

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Abstract
The scattering of visible light in optically dense sprays compromises the ability of common spray imaging techniques to capture the quantitative liquid mass distribution. Because of reduced susceptibility to multiple scattering, X-ray radiography can overcome the difficulties associated with optical wavelengths and provide quantitative measurements of liquid mass distribution in a variety of sprays. In the current work, we investigate the conditions under which broadband X-ray radiation from compact tube sources can be used to achieve similar accuracy to the narrowband, high-intensity, soft X-ray source at the Advanced Photon Source (APS) at Argonne National Laboratory. Radiography is used 2-D imaging of sprays with different liquid path lengths to evaluate the effects of energy dependent X-ray attenuation, or so-called beam hardening. We present 2-D imaging data using a time-resolved flash X-ray source. These measurements are compared with data acquired using the APS for the same spray conditions. The results indicate that quantitative images can be achieved using broadband X-ray sources, although the imaging system should be optimized to balance X-ray bandwidth, signal-to-noise ratio, sensitivity, spatial resolution, and temporal resolution.

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**Introduction**

Quantitative knowledge of multiphase flow phenomena is critical to a broad range of applications including chemical processing, drug delivery, material synthesis, power generation, and propulsion [1]. In the case of liquid fueled propulsion devices, the spray dynamics are coupled to combustion instabilities and potential rapid catastrophic failure. The measurement of liquid distribution and breakup are essential to the development of empirical and numerical models.

Qualitative optical techniques have been applied with good success in studies of jets in crossflow, aerated jets, and supercritical injection. These techniques include ballistic imaging, shadowgraphy, Mie scattering, holography, laser-induced fluorescence have been shown by [1-7]. Current quantitative techniques, based either on phase Doppler interferometry [8] or diffraction [9], are limited to far-field optically thin regions of the spray. Near-field spray imaging requires a technique capable of penetrating optically thick non-symmetrical regions.

X-ray radiography has been effective in imaging this dense near-field region. The dominant interaction of X-ray photons is absorption, while scattering remains relatively weak in comparison to visible light. The total signal attenuation can be related to the observed density along the line-of-sight, allowing for measurements of liquid mass distribution which are nearly independent of local spray geometry.

Leading work in X-ray spray radiography has emerged from the Advanced Photon Source (APS) at Argonne National Laboratory. The synchrotron source provides a tunable, highly collimated, low-energy beam with sufficient flux to image a wide range of path lengths and sufficient flux to obtain good temporal resolution. Because the narrowband beam passes through the sprays without a significant change in spectrum due to varying absorption, the attenuation coefficient remains nearly constant. Recent work in this area has been done by [10-17].

Radiography utilizing tube source X-rays has been used to show similar quantitative spray behavior. The broadband spectrum of tube sources results in higher energy photons (~100 keV) which increase photon scattering and the lower attenuation reduces image contrast. In general, the absorption and scattering of X-rays scale as the inverse cube and inverse square of photon energy, respectively. The broad spectrum leads to a wide range of photon attenuation coefficients. The preferential attenuation of low energy photons, or beam hardening, shifts the output spectrum and requires the use of a variable attenuation coefficient.

Application of polychromatic sources for investigation of instantaneous or time-averaged spray structure has been presented by a number of researchers, including [18-20], but only for qualitative imaging and without comprehensive comparison with narrowband sources.

Previous work at Iowa State demonstrated the ability to apply tube source radiography for sprays of practical interest by avoiding the effects of multiple scattering and preferential attenuation [21]. Tube sources are ideal for laboratory scale studies due to the small size and low capital investment. Tube sources have been used for time-averaged, two-dimensional radiography and three-dimensional computed tomography [1]. To the best of our knowledge, however, a direct validation of time-resolved liquid mass distributions measured using broadband X-ray sources has not been undertaken for atomizing sprays [22].

In the current work, measurements within atomizing sprays using a broadband flash X-ray tube source are compared with narrowband X-rays from the APS synchrotron facility. Time resolved liquid mass distributions measured from two-dimensional radiographs using the flash source are compared with time averaged raster-scanned profiles from the APS for different locations within the breakup and atomization regions. Because the APS can utilize lower energy X-rays, which are attenuated more greatly by the spray, thinner sprays may require the use of potassium iodide (KI) as a contrast enhancing agent. An impinging jet injector and gas-centered swirl-coaxial injector are used to investigate applicability to typical sprays in propulsion. Static measurements to investigate the potential effects of beam hardening for different KI concentrations in water [18-20] provide information on the feasibility, accuracy, challenges, and potential strategies for utilizing broadband X-ray tube in quantitative spray measurements.

**Experimental Setup**

The APS 7-BM beamline, a synchrotron bending magnet, was used to verify the accuracy of the flash images. This beamline provides a nearly collimated, polychromatic X-ray beam, and is housed in two enclosures. The first enclosure (7BM-A) contains slits to condition the beam size and a double multilayer monochromator (ΔE/E = 1.4%) to create a narrow beam. The monochromator has an energy range of 5.1–12 keV. The second enclosure (7BM-B) houses a pair of Kirkpatrick-Baez focusing mirrors [23], the experimental spray setup, and the X-ray detector. Further details regarding the beamline are given by Kastengren [24].

The current APS experiments use a focused X-ray beam with full width at half maximum (FWHM) dimensions of 5 μm (vertical) × 6 μm (horizontal) at 10 keV photon energy; with $1.6 \times 10^{10}$ photons per second
flux at the detector. The detector is an unbiased, 300-μm thick silicon PIN diode with 89% of the X-ray photons absorbed by the detector. The PIN diode output was amplified with a transimpedance amplifier and time-averaged over a 1 second integration time for each point. Data points collected from the raster-scanned spray were mapped to a two-dimensional grid. Points away from the spray were for background normalization. Each data was dark-current subtracted and flat-field normalized. The signal was converted to an equivalent path length (EPL) of liquid along the line of sight. Two-dimensional images were compiled via MATLAB. Because of the relatively small focal spot size, the spatial resolution of the images was limited by the raster scan spacing, which was a minimum of 50 μm near the jet centerline.

Obtaining temporal resolution in a two-dimensional radiograph using a flash X-ray source requires as much usable flux as possible. This poses new challenges to the emission and collection of radiation. Previous tube source time averaged studies using a metallic filter to preharden the beam greatly reduced the usable flux—a serious disadvantage for time resolved measurements.

![Flash X-ray Source](image)

**Figure 1.** Flash X-ray source experiment schematic (bird’s eye view) showing the geometrical effects of magnification and penumbra.

Flash X-ray pulses are produced when a bank of capacitors discharge in rapid succession and the full current pulse impinges upon the anode within tens of nanoseconds. The isotropic X-ray emission displays a bremsstrahlung spectrum with intense peaks characteristic of the anode material. The X-rays then pass through a thin beryllium window and the spray, and they are partially absorbed by the image plate shown in Fig. 1. The image plate is then read by a laser scanner and a digital image is formed which has been magnified and blurred. The unfiltered beam’s spectrum changes as it is attenuated through the spray, resulting in a varying attenuation coefficient. An approach is taken here to maximize the flux and model the attenuation coefficient.

The methods for optimizing the imaging system in these experiments are: source voltage, source anode size and material, source window material, anode-spray distance, spray-plate distance, image plate (detector), laser scanner, and system calibration. X-ray energies are tailored by selecting the number of capacitors used. Starting with a 150 keV system, half of the capacitors were removed to produce a 75 keV system resulting in a larger fraction of lower energy photons. The tubes in a flash source are readily interchangeable, and a 1 mm tungsten anode was chosen for longevity and focal spot size. Lower Z materials anodes can produce increased levels of lower energy photons, but these degrade with each pulse leading to inconsistent imaging behavior. A small focal spot size reduces the effect of penumbra, but will decrease the tubes useful life span. A beryllium window was chosen over steel, traditionally used, to reduce the attenuation of lower energy photons. The location of the anode, spray, and detector affects the total flux, the influence of penumbra, and the geometric magnification. The source is placed close to the image plate to increase flux. As shown in Fig. 1, if the spray is moved closer to the anode both the penumbra and magnification increase. The penumbra will increase at a greater rate than the magnification. When using a millimeter size source the penumbra may dominate any benefit of magnification so the spray was placed near the image plate. A Kodak GP image plate was chosen for contrast. The thickness of the plate determines the total absorbance and resolution. A thin plate tends to increase the resolution with a reduction in the total absorption. A ScanX HC laser scanner, with 50 μm focal spot size, was used to read the imaging plates. The spot size affects the resolution and contrast of the image, a smaller spot yielding greater resolution and lower contrast. Once the imaging system is designed, calibration images are taken and the system is modeled to determine the attenuation coefficient empirically and analytically. Liquid jets of known diameters were employed as quasi-static reference gauges to mimic a step wedge.
### Table 1. Measurement Parameters for X-ray Sources.

<table>
<thead>
<tr>
<th>Source</th>
<th>APS Synchrotron</th>
<th>Flash Tube Source</th>
</tr>
</thead>
<tbody>
<tr>
<td>Imaging Method</td>
<td>Raster Scan of Water</td>
<td>2-D Radiograph of Water</td>
</tr>
<tr>
<td>Beam Shape</td>
<td>Collimated</td>
<td>Cone</td>
</tr>
<tr>
<td>Spectrum</td>
<td>Monochromatic</td>
<td>Polychromatic</td>
</tr>
<tr>
<td>Energy [keV]</td>
<td>10</td>
<td>&lt; 75</td>
</tr>
<tr>
<td>Detector</td>
<td>PIN Diode</td>
<td>Laser Scanned Imaging Plate</td>
</tr>
<tr>
<td>Binning</td>
<td>None</td>
<td>3 x 3</td>
</tr>
<tr>
<td>Exposure [s]</td>
<td>1</td>
<td>2.00E-09</td>
</tr>
<tr>
<td>Pixel Size [µm²]</td>
<td>5 x 6</td>
<td>113 x 113</td>
</tr>
<tr>
<td>Time per Image [s]</td>
<td>~ 1000</td>
<td>~ 300</td>
</tr>
</tbody>
</table>

The calibration experiments were modeled in Matlab using the known or approximated behavior of system elements. The output spectrum was a linear approximation of a known 150 keV spectrum. The photon energy dependent attenuation coefficients were taken from NIST tables [25]. The imaging plate properties were provided by Kodak, and the laser scanner was assumed to be independent of photon energy [26].

It was found that the background in each image taken was not always consistent and traditional flat field normalization could not be used. The shape of the background was determined in a number of steps and approximated with a third degree polynomial. First each horizontal array was linearly normalized and pixel intensities significantly greater than the standard deviation were eliminated from the array. These intensities are believed to be within the spray; any weaker signals are assumed to be background signal. The array with signal removed is then fit with a polynomial. The polynomial fit is then used to normalize the original data array. The polynomial fit normalization can reduce the potential transmission error of 0.04 by half or more.

Table 1 compares and contrasts each of the sources and techniques. The APS has very high spatial resolution with a monochromatic beam. These features greatly reduce the potential measurement error at the APS. The flash technique can produce multiple, simultaneous, time-resolved, two-dimensional images, but certain challenges must be overcome.

The narrow band X-ray beam from the APS allows for a constant attenuation coefficient to be used to calculate the equivalent liquid path length (EPL) from a simplified version of Beer’s Law, Eq. 1 and 2. Where $\alpha$ is the attenuation coefficient determined from attenuation cross section, $\sigma$, and number density, $N$.

\[
\frac{I(y,z)}{I_0(y,z)} = e^{-\alpha EPL} \quad (1) \\
\alpha = \sigma N \quad (2)
\]

When using a tube source the attenuation coefficient’s dependence on wavelength becomes prevalent. The relationship between the intensity and the EPL is now dependent on the convolution of energy dispersion and the detector response function, $\psi$, Eq. 3. The dependences are combined in the attenuation coefficient described as a linear function, dependent upon intensity with coefficients $m$ and $b$, Eq. 4.

\[
\frac{I(y,z)}{I_0(y,z)} = \int e^{-\int a(\lambda,y,x,z)dx \psi(\lambda)d\lambda} \quad (3) \\
\frac{I(y,z)}{I_0(y,z)} = e^{-\left[m\left(\frac{1}{I_0}\right)+b\right]EPL} \quad (4)
\]

The attenuation coefficient was modeled using known values for the source, spray, and detector. The 75 keV spectrum was approximated from an empirical 150 keV spectrum, using the same source at increased voltage, with a linear correction factor shown in Fig. 2. The 150 keV spectrum is multiplied by the linear correction factor to rescale the intensities while maintaining the approximate shape and characteristic peaks and edges. The spectrum is only used to estimate the general behavior of the attenuation coefficient.
Using the approximated 75 keV spectrum three spectrums are modeled: an incident spectrum, a spectrum after the X-rays pass through 2 mm of water and a spectrum after the X-rays pass through 2 mm of 15% KI. NIST attenuation tables are used to model the attenuation by the tube source window, the air, and the water or KI solution. Data from Kodak was used to model the imaging plate absorption and photoemission. It was assumed that the reading by the laser was independent of the X-ray spectrum.

The incident spectrum is greatly attenuated by the window and air alone. Figure 3 shows the preferential attenuation as seen by the large decrease in lower energy intensities relative to higher energy intensities, most noticeably in the case with 15% by mass KI.

Two injectors were imaged to compare the flash source and the APS. The first is a like-doublet impinging-jet injector with an included angle of 60 degrees, as shown in Fig. 4. The injector orifices have a diameter of 0.51 mm and length-to-diameter ratios of 46 with chamfered inlets.

A gear pump drives the liquid at impinging jet velocities of 3.5 and 8 m/s. Shown in Fig. 5 are shadowgraphs of the impinging jet sprays with the two jets approaching the impingement zone in the plane perpendicular to the plane of the image. The jets meet roughly 3 mm below the injector tips. The 3.5 m/s spray forms a semi-stable liquid sheet with most of the liquid occurring in the rim. The unstable 8 m/s spray forms a wavy sheet with disintegrating rim.

The second spray of interest is a gas-centered swirl coaxial injector, Fig. 6. Four liquid jets circle a gaseous jet and, in the case of incomplete breakup, the liquid jets are entrained in the gaseous flow and form irregular liquid ligands. The air flow rate was 227 lpm and the water flow rate was 1.9 lpm.
Figure 6. Gas-centered swirl coaxial injector schematic.

Shown in Fig. 7 is a shadowgraph of the gas-centered swirl coaxial injector spray. The transient asymmetrical structures require a technique that can penetrate a dense droplet field. A time-resolved two-dimensional image distinguishing the core structures becomes necessary.

Information on the liquid mass distribution is very difficult to discern in shadowgraphs, however with high spatial and temporal resolution it is possible to capture some characteristic external features of the spray.

Figure 7. Shadowgraph of the gas-centered swirl coaxial injector spray used for simultaneous stereography.

Results and Discussion

Correction of the attenuation coefficient for beam hardening is crucial for measurement accuracy. To capture the behavior of the coefficient over a range of path lengths, a laminar liquid jet study was conducted. Sixteen different jet diameters are used similarly to using a step wedge. A coefficient is found for each jet so that the experimental volume matched the theoretical volume of the jet. This coefficient was then plotted against the average path length to visualize the behavior in Figs. 8 and 9 and plotted against average transmission in Figs. 10 and 11 to determine the attenuation coefficient with a linear fit. This linear relationship is used as the attenuation coefficient to calculate the EPL in unknown sprays of interest. This approach presumes a linear coefficient, so that the average EPL of each jet is representative of the signal, while the model displays a curvilinear coefficient. The model was used to estimate the error of this assumption and it was found that the model overestimated the coefficient by less than 1.5% for path lengths less than 2 mm. This does not account for a majority of the error most likely due to the approximated spectrum of the flash source.

Figure 8. Plot comparing the empirical and modeled attenuation coefficients vs. EPL for water.

Figure 9. Plot comparing the empirical and modeled attenuation coefficients vs. EPL for 15% KI.
The linear fit has an RMS value of 0.13 and 0.60 for water and 15% KI respectively. This can be due to any number of factors including but not limited to noise, changes in tube anode output, and changes in imaging plate behavior.

The model of the attenuation coefficient appears to agree well with the data, within 13% and 11% on average for path lengths less than 2 mm. Due to the amount of noise in the current measurements, this curvilinear behavior could not be captured nor would affect the end result appreciably beyond the current uncertainty. The uncertainty in the model has not yet been determined. The agreement does show the dominant physics are being captured. The largest error in the model is likely from the approximated input spectrum.

Table 2 compares the imaging method capabilities of the X-ray sources. The APS images are of higher contrast, spatial resolution, and accuracy. Despite their lower fidelity, the flash images are capable of quantitative time-resolved 2-D imaging. Differences in spatial resolution in the flash X-ray technique are due to the physical location of the source, spray, and detector. The differences in noise (EPL std. dev.) and potential error are due the different liquids used.

In the following discussion time resolved flash radiographs are compared against time averaged APS measurements and two flash sources are paired orthogonally to increase the spatial information of the imaging technique. The impinging jets used 15% KI and the anode-spray distance was 200 mm and the spray-detector distance was 25 mm. The gas-centered swirl coaxial injector sprayed water and the anode-spray distance was 250 mm and the spray-detector distance was 63 mm.

Impinging jets were imaged using the flash source with 15% KI, as in Fig. 12. The impact waves traveling down the center the liquid sheet are plotted against the time averaged water data from the APS, as shown in Figs. 13-14. Impact waves form in both the high and low flow cases are always apparent in the 8 m/s spray and much less frequent a 3.5 m/s. The flash data follows the APS trend quite well, and the instantaneous 2-D capability enables detection of these impact waves.

The standard deviation of the APS data is 0.3 µm where the flash data standard deviation is 25 µm. At 2 mm where the jets are first meeting the lower spatial resolution of the flash radiographs do not capture the exact average shape.

The spatial resolution was estimated to be 350 µm at a 10-90% rise criterion. When the spray is thicker and less symmetric, stereography using water can be employed.
of 450 µm 10-90% rise distance was sufficient to capture the large intact structures of interest. The orthogonal radiographs show the severe asymmetry of the spray. These highly transient structures are not easily detected in time averaged images where the movements average out the spatial differences.

Figure 12. Flash radiographs of impinging jet sprays of 15% KI at 3.5 m/s (left) and 8 m/s (right) jet exit velocities.

Figure 13. Line plot down the center of the 3.5 m/s impinging jet spray for the APS and flash tube source data.

Figure 9. Line plot along center of the 8 m/s impinging jet spray for the APS and flash tube source data.

Figure 10. Simultaneous orthogonal flash radiographs of gas-centered swirl coaxial injector spray.

Figure 16 shows a line plot 3 mm below the injector face. The plot agrees with time averaged data previously taken by [17]. The peaks in EPL represent where the intact liquid structures are occurring. This data set is at the lower limit of the current imaging systems capabilities. The standard deviation of the non-binned data is equivalent to an EPL of 100 um; the plot is binned 3 by 3 but still susceptible to errors in the normalization and attenuation coefficient. The total volume in each line plot should be equal but differs by 30%. In addition to previously mentioned errors, the second tube source was not calibrated. Hence, some disagreement is not unexpected. Nonetheless, it is possible to associate quantitative meaning to regions with low and high absorption for the same tube source. The end result is an improved understanding of ligand structure sizes, locations, and liquid mass distribution.

Figure 11. Line plots of gas-centered swirl coaxial injector spray taken 3 mm below the injector face.
Summary

The current work examined the applicability of flash X-ray sources as quantitative means of determining liquid mass distribution in sprays. The comparison with the APS shows these measurements are possible with proper attention paid to image processing. Further comparisons with time-resolved one-dimensional imaging at APS would further establish the abilities and usefulness of flash X-ray radiography and stereography.

In future experiments, higher fidelity can be achieved by collecting a larger number of calibration images, and calibration should be conducted for each source and detector. The simultaneous calibration would allow for minor changes in attenuation coefficient to be accounted for on a shot to shot basis. The normalization may also be improved by taking a larger number of background images and determining a general trend to help decrease the error.

Empirically determining the actual source spectrum would increase the fidelity of the model and this knowledge may also aide in the calibration and determination of the variable attenuation coefficient.

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