

# The Effects of Dissolved Methane upon Liquid Argon Scintillation Light

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**ABSTRACT:** In this paper we report on measurements of the effects of dissolved methane upon argon scintillation light. We monitor the light yield from an alpha source held 20 cm from a cryogenic photomultiplier tube (PMT) assembly as methane is injected into a high-purity liquid argon volume. We observe significant suppression of the scintillation light yield by dissolved methane at the 10 part per billion (ppb) level. By examining the late scintillation light time constant, we determine that this loss is caused by an absorption process and also see some evidence of methane-induced scintillation quenching at higher concentrations (50-100 ppb). Using a second PMT assembly we look for visible re-emission features from the dissolved methane which have been reported in gas-phase argon methane mixtures, and we find no evidence of visible re-emission from liquid-phase argon methane mixtures at concentrations between 10 ppb and 0.1%.

**KEYWORDS:** Noble-liquid detectors; Photon detectors for UV, visible and IR photons; Scintillators, scintillation and light emission processes.

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## 1. Motivations for Studying Methane in Liquid Argon

Many current- and future-generation neutrino and dark matter experiments use liquid argon as an active medium [1–7]. The detection of weakly interacting particles-of-interest in such a detector involves collecting ionization charge and/or scintillation light produced in the argon bulk. In order to achieve a large target mass with a reasonable number of sensitive detector elements, the detection of both light and charge typically takes place some distance from the interaction point. For charge collection, this is achieved by drifting free electrons in an electric field, as in a time projection chamber (TPC). For light detection, sensitive elements such as PMTs [8] or light guides [9] are placed around the boundaries of an argon target volume. Because argon is transparent to its own scintillation light, some fraction of the isotropically produced scintillation light is captured by these sensitive elements.

Both charge and light production and propagation are known to be affected adversely by small concentrations of dissolved impurities. The commercially available argon which is used in most liquid argon detectors is manufactured by the distillation of air, and typically arrives from the vendor with part-per-million (ppm) levels of contaminants such as water, oxygen and nitrogen [10]. For a TPC detector, water and oxygen must be removed in a subsequent filtering process to the hundred parts per trillion level in order not to damage the free electron lifetime [11]. In a scintillation detector, not only water and oxygen, but also nitrogen must be controlled at the ppm level in order to prevent scintillation quenching [12–14] and absorption [15].

A new generation of dark matter experiments, which require highly radiopure argon [7], plan to use argon that is not distilled from air, but rather Underground Argon (UAr) extracted from underground sources. This argon has the advantage of having low levels of the radioactive isotope  $^{39}\text{Ar}$  [16], a source of background in such detectors. The UAr is extracted from carbon dioxide wells that are comprised of 96% carbon dioxide and 2% nitrogen, alongside sub-percent levels of

other gasses, including 5,700 ppm methane and 600 ppm argon. Locally at the CO<sub>2</sub> well, an Ar-He-N<sub>2</sub> mixture is extracted from the crude CO<sub>2</sub> gas, with methane levels below the sensitivity on the *in situ* gas analyzer [17]. The gas mixture is sent to the Fermi National Accelerator Laboratory for further purification, where, after He removal, cryogenic distillation removes the N<sub>2</sub> and purified UAr is produced [18]. This cryogenic distillation process not only concentrates the UAr, but also has the effect of concentrating any residual trace amounts of methane. It is therefore critical to know what concentration of methane is permissible in the UAr scintillation experiments in order to understand what purification techniques are required to remove the methane, which can be technically difficult.

On the other hand, mixed methane and liquid argon TPCs have been proposed as detectors for low-energy neutrino physics [19]. The benefit provided by the methane admixture is that free protons are present in the detector as interaction targets. This reduces the threshold for the interactions of low-energy neutrinos, such as those from the sun, or from supernovae, or man-made neutrinos from decay-at-rest sources. Free protons may also make neutron captures detectable in a liquid argon scintillation detector, allowing for coincidence tagging of low-energy antineutrino inverse-beta-decay interactions ( $\bar{\nu}_e + p \rightarrow e^+ + n$ ).

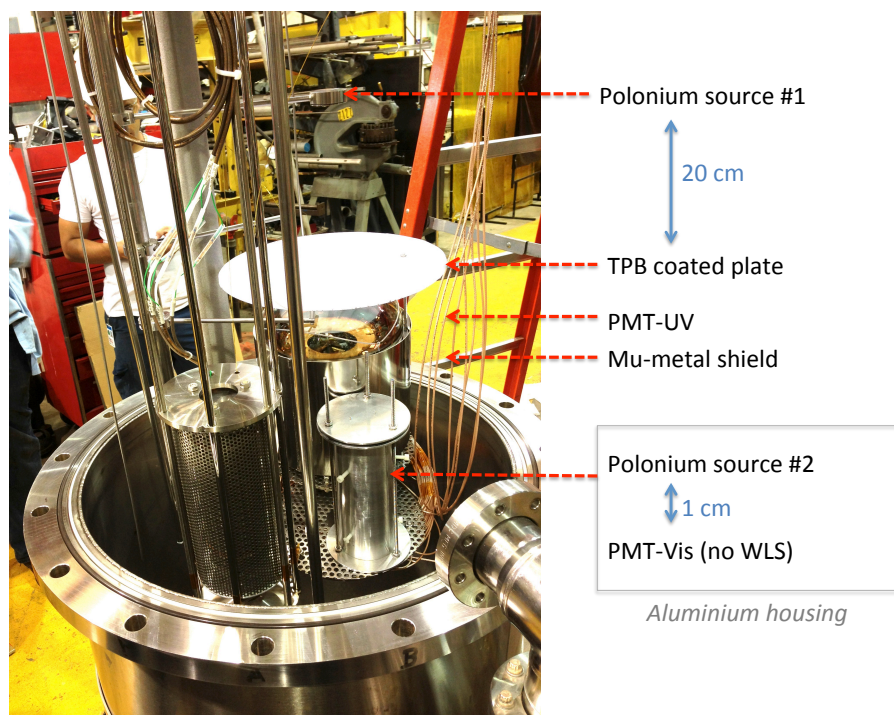
The effects of methane upon charge collection have been shown to be minimal, even at few-percent level concentrations [19]. However, the effects of methane upon liquid argon scintillation light have not been investigated previously. As such, no specification on the allowed methane concentration for a scintillation detector currently exists. World data on the VUV absorption cross section of pure methane, taken at room temperature and pressure, suggest that significant absorption may be expected at relatively low concentration levels [20]. Studies of scintillation light from gaseous argon-methane mixtures, however, have reported a visible emission feature at a wavelength of a 431 nm [21, 22]. This motivates a determination of the effects of methane impurities in liquid argon detectors, operating under realistic scintillation detection conditions.

In this paper we examine the effects of methane upon the light yield from alpha particles detected in a liquid argon scintillation detector, and quantify the concentration scale to which methane must be removed in order to prevent significant scintillation light losses relative to pure argon.

## 2. Experimental Configuration for this Study

These tests were performed using the Bo cryostat and gas injection system, which was described in detail in [15]. A polonium disc alpha source is held 20 cm from a cryogenic PMT assembly, which is comprised of a TPB coated wavelength shifting plate supported above an 8-inch cryogenic Hamamatsu R5912-02mod PMT [8], mounted inside a mu-metal shield. This is referred to henceforth as PMT-UV. Signal and high voltage are carried by a common RG-316 type coaxial cable, and split outside of the cryostat by a HV splitter unit. The signal is connected to a Tektronix DPO5000 oscilloscope terminated at 50Ω and sampling at 1 gigasample per second. Further details about this part of the system can be found in [15].

In addition to the apparatus described in [15], a 2-inch R7725-mod PMT, housed inside an aluminum cylinder, was installed for this test. This PMT will be referred to as PMT-Vis. The 2-inch PMT has no accompanying wavelength shifter and is shielded from visible light generated by the PMT-UV wavelength shifting plate. A second polonium disc alpha source is held 1 cm

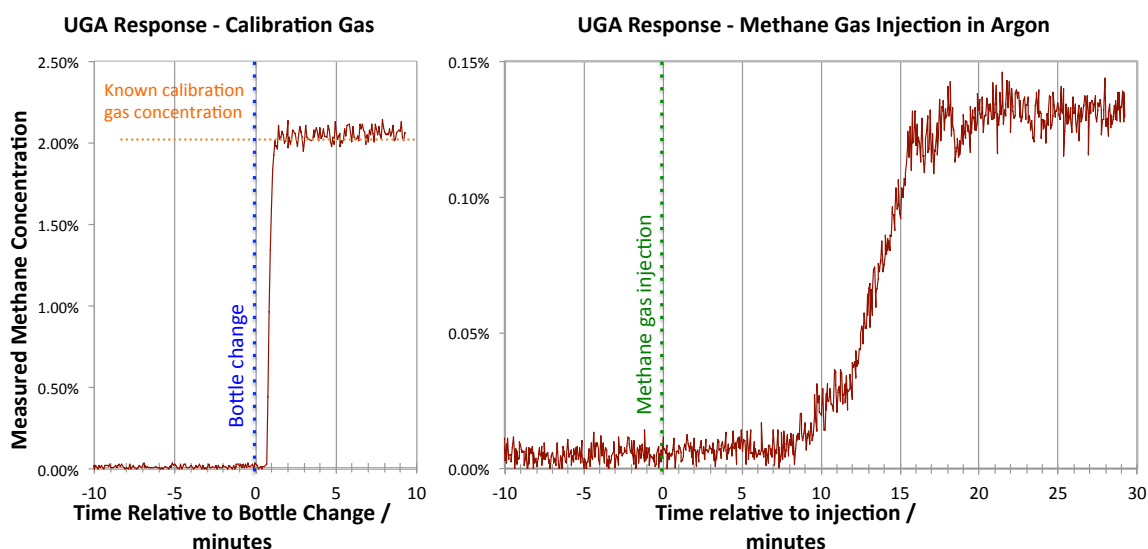


**Figure 1.** A labeled photograph showing the internal Bo configuration used in this study

from the face of the PMT-Vis. Openings in the base and around the top edge of the aluminium cylinder provide space for argon to flow through the two inch assembly to ensure that there is mixing throughout the system. A labeled photograph of the entire apparatus inside the cryostat is shown in Figure 1.

Our measurement involves making injections of small amounts of methane gas into the liquid argon volume, and studying the light yield at both PMT-UV and PMT-Vis. The light yield at PMT-UV indicates to what extent the 128 nm argon scintillation light has been absorbed or quenched by the injected methane. It would also be sensitive to visible re-emission if any were present. The light yield at PMT-Vis is sensitive to visible re-emission features only, and the fact that there is such a short distance between the alpha source and the PMT face means that this PMT should be sensitive to very low yields of visible light, on the order of 10 photons per MeV.

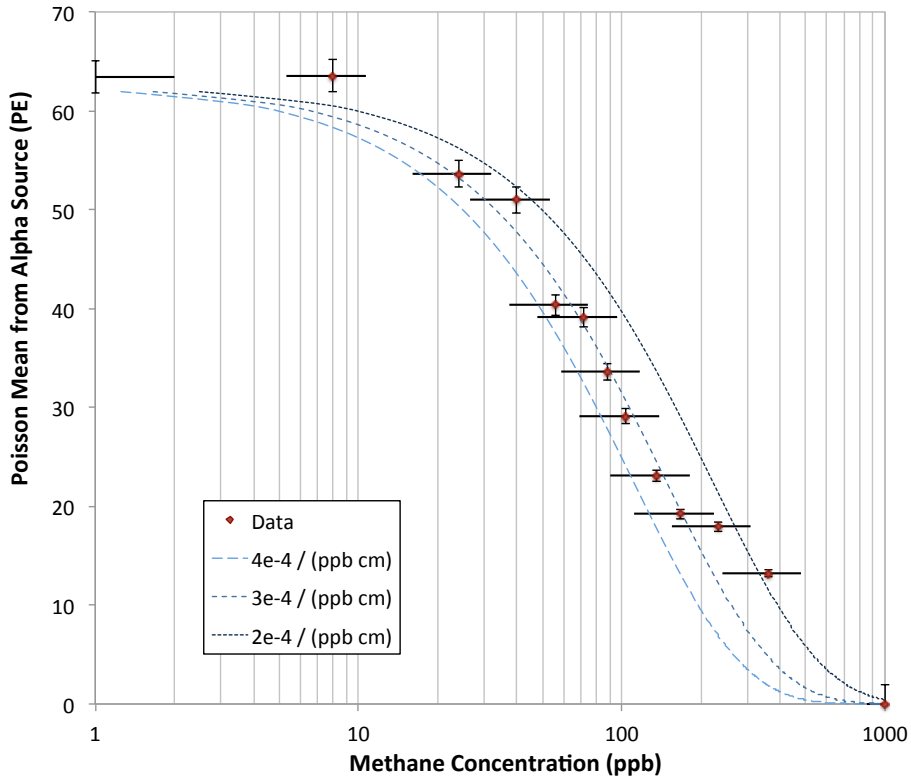
The equilibration time of methane in argon following an injection in this system determines the amount of time which must be left between methane injection and light measurement. We measured the equilibration time of the system after a methane injection in a preliminary study using a universal gas analyzer (UGA) [23]. First, the gas analyzer was connected to the Bo liquid sample line with the cryostat held at 12 psig. A stable methane concentration at the noise floor of the device, corresponding to around 0.01% methane by mole, was observed. At a known time, the UGA piping was disconnected from the Bo cryostat and connected to a bottle of 2% methane-in-argon calibration gas regulated at 12 psig. The time taken for the methane concentration reported by the UGA to stabilize to 2% was approximately one minute, which sets the timescale for flow of gas



**Figure 2.** Left: Time taken for reported methane concentration by UGA to stabilize after swapping source from Bo sample line to calibration gas at the Bo cryostat location. This quantifies the flow time in the sample piping and the lag time of the UGA. Right: Time taken for reported methane concentration by UGA to stabilize after an injection of methane into pure argon in the Bo cryostat. The equilibration time is approximately 20 minutes.

through the 30 m supply pipe and the lag time of the measuring device. Then, with Bo reconnected and the methane reading stable at the noise floor once again, an injection of around 0.1% methane was made into Bo. The reported methane concentration by the UGA is seen to rise gradually, and then stabilize after approximately 20 minutes. Since we already know that the flow time in the piping and the UGA response time is much shorter than this, we conclude that 20 minutes is the equilibration time for methane injected into the cryostat to reach an equilibrium distribution between the gas and the liquid phases. These measurements can be seen in Figure 2. We allow a conservative 40 minutes of equilibration time between any injection and light yield measurement. The concentrations of methane we investigate in this paper are far below the sensitivity of the UGA, and so we cannot use it to quantify the injected methane. Our only concentration measurements come from the known volume and pressure of the injected gas, which is accompanied by relatively large systematic uncertainties. We describe these below.

For the tests described in this paper, the 56 cm diameter cryostat was filled to a level of  $76 \pm 1$  cm with high-purity liquid argon, and maintained at a pressure of  $12 \pm 0.2$  psig by a liquid nitrogen condenser system. In order to achieve ppb level concentrations of methane in argon, we inject a pre-mixed gas of 2% methane in argon via a  $300 \text{ cm}^3$  injection canister, which can be charged to a known pressure with gas which is then released into the liquid through an injection capillary. To achieve injected methane concentrations at the 10 ppb level, the injection pressure must be set fairly low relative to the design pressure of the injection system. The lowest methane concentration point requires a 15 psig injection into the 12 psig Bo volume, so the pressure gauge uncertainty of 1 psig implies a 33% systematic error on the internal methane concentration. Since we are continually adding methane to the same argon volume, the systematic error on the methane

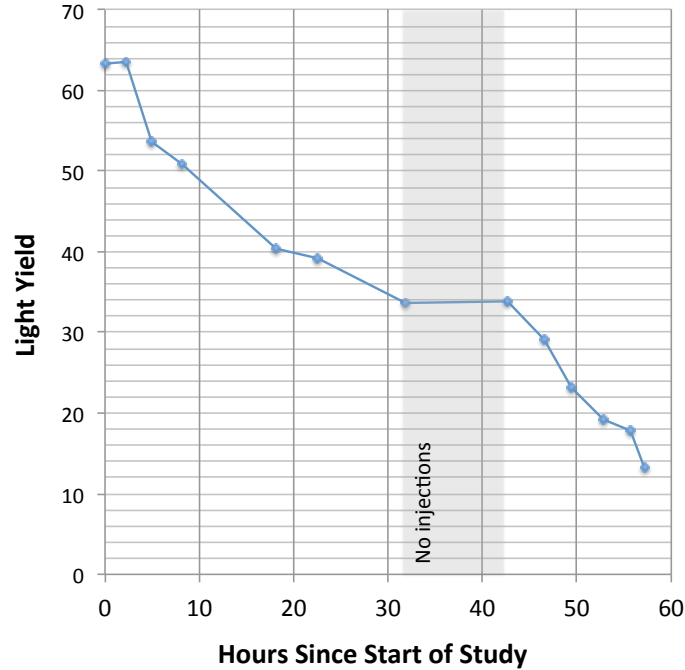


**Figure 3.** The measured prompt scintillation light yield from a monoenergetic polonium 210 alpha source as a function of methane concentration. Horizontal error bars are given by the precision of the injection pressure gauge. Vertical error bars are given by the single photoelectron scale added in quadrature with the peak fit error. The left-most point is for pure argon. The overlaid curves show the predicted attenuation behavior based on a ray tracing simulation for three characteristic attenuation strengths.

concentrations for subsequent concentration points are highly correlated. For future higher precision studies, the injection system may be improved by installing a smaller injection canister or higher precision pressure gauge. For the purposes of this paper, however, a logarithmic scan of the relevant concentration range is already very instructive.

### 3. The Effect of Methane upon Prompt Scintillation Light Yield

In order to study the effects of injected methane upon the prompt scintillation light yield, we follow a very similar procedure to the one described in [15]. We collect 40,000 pulse areas within a narrow window around a falling-edge trigger from PMT-UV and histogram these online using the oscilloscope. We fit the pulse area distribution with a function describing a poisson-like peak with corrections for source shadowing added to a falling power-law background. Many more details of this procedure can be found in [15], Section 3. The average DC baseline is measured and subtracted using the pre-samples for 10,000 collected single waveforms, and the single photoelectron (SPE) scale is measured at several points throughout the run using late scintillation light. For this run, the measured SPE was found to be stable to within 2.5%. The light yield at every concentration



**Figure 4.** The prompt light yield as a function of time during this study. For the period where no methane was injected, the light yield was found to be stable to within 1.2%

point is normalized to the most recently measured SPE scale, with the most recently measured DC baseline subtracted.

The alpha-induced poisson-like distribution mean is extracted as a function of the methane concentration. These data are shown in Figure 3. We see no observable light loss below 10 ppb, and then steady losses of scintillation light between 10 ppb and 1 ppm. Beyond 1ppm of methane, the alpha peak was no longer observable.

Overlaid on the figure are the predictions of the expected light yield as a function of methane concentration for absorption strengths of 0.02% , 0.03% and 0.04%  $\text{ppb}^{-1}\text{cm}^{-1}$  as determined by a ray tracing simulation of our apparatus. A similar ray tracing model was used in order to extract the absorption strength of nitrogen in [15]. These correspond to molecular absorption cross sections of  $9.5 \times 10^{-18} \text{ cm}^2$ ,  $1.4 \times 10^{-17} \text{ cm}^2$  and  $1.9 \times 10^{-17} \text{ cm}^2$  respectively. Our data are in approximate agreement with previous measurements of the methane absorption cross section at 128 nm, made at room temperature and pressure with pure methane gas [20]. We address in more detail whether the losses we see can be attributed to absorption effects alone in section 4.

As in [15], as a cross-check that the light losses observed were a result of methane injection and not outgassing of water or some other system instability, we collected similar datasets without injecting methane 12 hours apart during the middle of the run, and found stability of the light yield to within 1.2%. This is to be contrasted with the tens-of-percent light losses observed over 12 hour periods during methane injections. These data are shown in Figure 4. In a separate run, a single methane injection of 1ppm was made and the alpha-induced poisson-like peak was immediately extinguished.

#### 4. Argon Scintillation Time Constants, Quenching and Absorption

The light losses described in section 3 could be the result of quenching, absorption or both. In this context, quenching refers to a process where the excimers involved in the liquid argon scintillation process are dissociated by interaction with an impurity molecule without emitting a scintillation photon. This mechanism affects late, triplet-state scintillation light much more strongly than prompt, singlet-state light, leading to a reduced time constant and late-to-total light ratio. Absorption, on the other hand, refers to the loss of photons due to interactions with impurities during propagation between source and detection points, and affects both prompt and slow scintillation light equivalently.

To distinguish between these two types of processes we investigate the effects of methane concentration upon the time constant of the late liquid argon scintillation light. If the large methane-induced losses observed in the prompt scintillation light are a result of a quenching process, we would expect large changes in the slow scintillation time constant at concentrations at or below those where the prompt peak is affected. A pure absorption process would affect both components equally, leading to an overall reduction in light yield with no change of time constant.

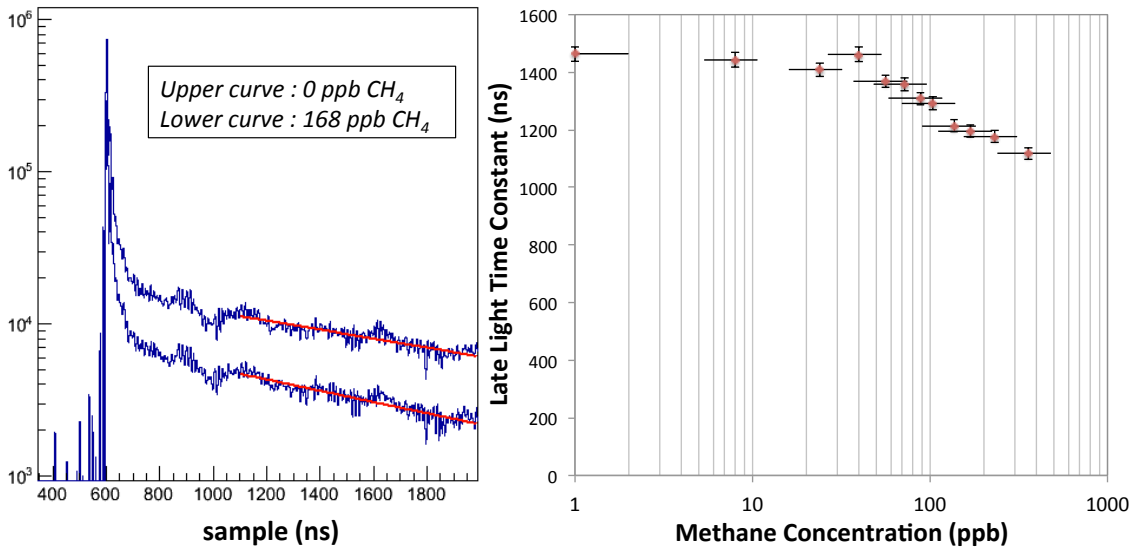
The method used to extract the late scintillation time constant resembles the method reported in [12]. The single photoelectron response shape was first measured using a pulsed LED, optically coupled to a fiber which points at the PMT-UV photocathode. Then, 10,000 self-triggered and baseline-subtracted waveforms were summed to produce an average PMT-UV pulse shape for scintillation deposits at each concentration point. The summed waveform undergoes some pre-processing to prevent ringing artifacts from deconvolution, including the application of a smooth sigmoid window function at the sample edges, and pre- and post-padding with zeroes to prevent wraparound effects. The summed waveform is then deconvolved using the single photoelectron pulse shape as a deconvolution kernel, and an optimized Weiner filter combined with low-pass filter is applied to suppress noise and deconvolution artifacts.

This process removes all linear shape effects due to the PMT anode response, PMT undershoot, ringing in the PMT base electronics, and reflections in cables. Nonlinear contributions to the pulse shape are not removed by this method. As such, the reported late-light time constant obtained from this method is valid only to the extent that the PMT and electronics response is linear. The level of system nonlinearity is very difficult to quantify. For the purposes of this study, we assume that the system response is linear and thus the measured time constants are faithful. Since our primary focus is upon changes in the late scintillation time constant rather than its absolute value, our main conclusions remain valid even in the presence of small nonlinear shape effects.

To extract the late-light time constant we fit the deconvolved pulse shape from 500 ns after the prompt pulse with a falling exponential. The 500 ns delay is chosen to avoid the effects of afterpulsing, deconvolution artifacts from the sharp edge of the prompt peak and the effects of any possible intermediate scintillation time constants. The measured PMT dark rate is sufficiently low that no constant offset term is required in the exponential fit. Two deconvolved average waveforms and their corresponding fitted exponential functions are shown in Figure 5, left. The dependence of the measured time constant on methane concentration is shown in Figure 5, right.

As has been discussed in appendix C of [12], different definitions of the argon scintillation time constants and different fit methods used to extract them from deconvolved pulses can lead



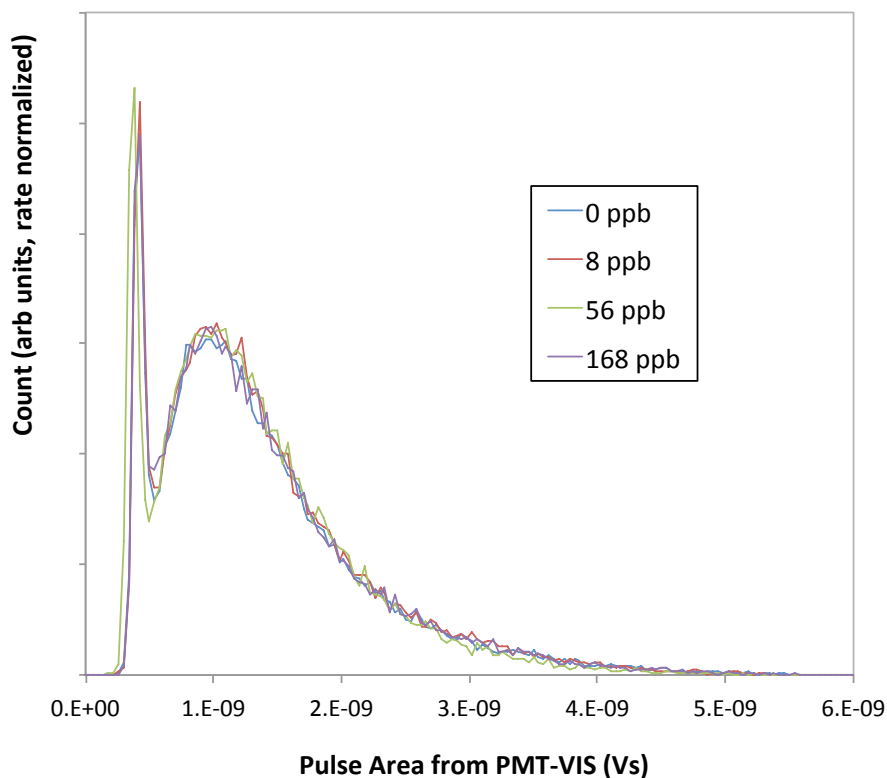


**Figure 5.** Left : Average deconvolved pulse shapes and exponential fits for two concentration points. Right: The measured late light time constant vs methane concentration. Vertical error bars are given by the RMS discrepancy between fitting the first and second half of each dataset. The left-most point is for clean argon.

to different numerical values. By fitting exponentials to shorter sections of the deconvolved pulse we were able to obtain time constants between 1450 and 1630 ns for clean argon. However, the relative variation of the time constant as a function of methane concentration was found to be unchanged within systematic errors. In this study we are interested in this variation, rather than the absolute value of the time constant. For this reason, the error bars shown in Figure 5 do not include this correlated systematic uncertainty, and are determined instead by the discrepancy between the time constants obtained from independently fitting the first and second half of the dataset for each concentration point, with the fit performed over the full fit region.

These data show very clearly that the majority of the light losses observed in the prompt scintillation light yield are not the result of a quenching process. The late scintillation time constant remains long even when significant prompt light losses are observed. Some evidence of a quenching process is observed at higher concentration values, but this effect is much too small to explain the observed light losses in the prompt peak. The two curves of Figure 5 are for an unquenched and a heavily quenched point from this study. The change in time constant between the 0 and 168 ppb concentration points is relatively small, whereas a significant loss of light yield for both prompt and late light components is clearly visible.

We do not have the ability to test this apparent quenching behavior at much higher concentrations of methane in this detector, since the absorption effect is dominant and blocks a large fraction of the light required to observe the quenching. A detailed study of this quenching process would be ideally performed in a much smaller detector, where light losses due to absorption between source and detector are smaller. However, for any realistic scale neutrino or dark matter detector, the absorption effect is likely to be the dominant mechanism of light loss.



**Figure 6.** The rate-normalized charge per pulse measured on PMT-Vis, showing pedestal and SPE. No SPE rate increase or higher intensity visible emission feature is observed as methane is injected.

## 5. Searching for Visible Re-emission Features

It has been reported that a visible scintillation emission feature has been observed in gas-phase mixed argon and methane drift chambers [21]. The feature is suggested to be a 431 nm emission line resulting from interactions between argon excimers and various energy levels of methane molecules [22]. A competing non-radiative decay route at high methane densities is suggested to quench the emission, which is likely to suppress any such feature in a liquid phase mixture, where the molecular mean-free-path is much smaller than in the gas phase. However, if some visible emission feature is present despite the strong absorption of 128 nm scintillation light, it could help to circumvent the problems associated with the absorption by allowing visible light to propagate across the detector, unabsorbed by methane.

We search for visible emission features by measuring the areas of self-triggered PMT signals from PMT-Vis within a narrow window of a falling-edge trigger. This PMT is optically isolated from most of the argon volume, but has a polonium disc source held 1 cm from the PMT face. In pure argon, no features larger than the single photoelectron background peak are observed.

The single photoelectron dark rate in clean argon is comparable to the alpha source rate; they are around 200 Hz and 100 Hz respectively. This means that even triggering at the single photoelectron level, any alpha induced visible emission feature would be observable over background.

Rate-normalized histograms of pulse areas for several methane concentration points are shown in Figure 6. No change in the single photoelectron rate is observed, and no higher intensity features are detected for any concentration point. These data were taken up to 168 ppb of methane, by which point 70% of the 128 nm scintillation light was being absorbed. We also performed a logarithmic scan of concentration points between 1 ppm and 0.1% methane by mole, none of which have any observable 128 nm scintillation light, and saw no evidence of strong visible re-emission features at any concentration.

## 6. Implications for Neutrino and Dark Matter Experiments

Our results show that significant light loss is to be expected when as little as tens of ppb of methane are mixed into liquid argon, due to ultraviolet absorption of 128 nm photons. The loss of light is accompanied by no detectable visible re-emission, and as such, this poses a problem for any scintillation detectors operating in this methane concentration range.

In the case of dark matter detectors using UAr, to eliminate the adverse affect of methane on argon scintillation, it is clear that the argon must contain less than 10 ppb of methane. To reach this goal, hot getters can be used [24]. Since the light losses are attributed to an absorption process, the exact purity specification will depend on the geometry of the detector in question, with larger detectors requiring higher purity.

For TPC detectors aiming to use argon/methane mixtures in order to provide free protons, the detection of scintillation light is likely to be impracticable. In order to provide a reasonable number of free protons for their physics goals, these detectors would need to contain methane at the few percent level or higher. Whilst this type of argon/methane mixture has been shown to have acceptable properties for electron drift and charge collection [19], our studies show that such a detector would be blind to scintillation light, because of the methane absorption effect.

At higher concentrations, we have also seen evidence of an apparent quenching effect, which is inferred from a reduction of the late scintillation light time constant. The configuration of our apparatus described in this paper is not well suited to studying the quenching effect in detail, since by the time the effect is large, most of the emitted light has been absorbed between source and detector. This is likely also to be true of any realistic neutrino or dark matter experiment, and so the practical implications of this quenching effect are limited. A detailed study of the effect could be made in a test with a much smaller source to PMT path length, and with a data acquisition system capable of recording longer waveforms in order to capture many lifetimes of late scintillation light.

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