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1	Under-expanded jets and dispersion in high pressure CO ₂
2	releases from a large-scale pipeline
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4	Xiaolu Guo ^a , Xingging Yan ^a , Jianliang Yu ^{a†} , Yongchun Zhang ^b , Shaoyun Chen ^b ,
5	Haroun Mahgerefteh [°] , Sergey Martynov [°] , Alexander Collard [°] , Christophe Proust ^d
6	
7	^a School of Chemical Machinery and Safety,
8	Dalian University of Technology, Dalian, 116024, China
9	
10 11	Dalian University of Technology Dalian 116024 China
12	Danan Oniversity of Teenhology, Danan, 110024, China
13	^c Department of Chemical Engineering,
14	University College London, London WC1E 7JE, UK
15	damara a materia da servicia
16	^a INERIS, Parc Technologique ALATA, Normauil an Halatta BB 2, 60550, France
17	Verneun-en-Halatte BP 2, 00330, France
18	
19	
20	
21	
22	[†] Corresponding Author
23	School of Chemical Machinery and Safety
24	Dalian University of Technology
25	No 2 Ling Gong Road
26	Dalian, 116024,China
27	e-mail: yujianliang@dlut.edu.cn
28	Tel.: +86 411 84986281
29	Fax: +86 13998576027
30	
31	
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Under-expanded jets and dispersion in high pressure CO₂ releases from a large-scale pipeline

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Xiaolu Guo^a, Xingqing Yan^a, Jianliang Yu^{a†}, Yongchun Zhang^b, Shaoyun Chen^b, Haroun Mahgerefteh^c, Sergey Martynov^c, Alexander Collard^c, Christophe Proust^d

Abstract

8 The widespread implementation of Carbon Capture and Storage (CCS) in industry will 9 require extensive long-distance CO₂ pipeline networks to integrate the component technologies. The potential for pipeline rupture and leakage, possibly resulting in catastrophic 10 accidents, will inevitably increase as networks become more extensive. The study of 11 near-field source terms and dispersion behavior after pipeline rupture is an essential 12 13 foundation of CO₂ pipeline risk assessment and will provide effective technical support for 14 the implementation of large-scale CCS projects and contribute to pipeline safety. In the CO₂QUEST project under-expanded CO₂ jets, cloud dispersion characteristics and the 15 formation of dry ice particles in the near field were investigated during releases from a 258 m 16 long, fully instrumented pipeline. Experimental data including cloud temperature, CO2 17 concentration and the visual evolution of the cloud (recorded on film), was gathered to 18 19 investigate cloud behavior and to support future work in the field of CO₂ pipeline safety. 20 Experiments included the release of gaseous and dense phase CO₂ through three orifice 21 diameters: 15 mm, 50 mm and Full Bore Rupture (FBR). The lower limit of gaseous CO₂ concentration for adverse effects in humans is 5 % v/v. Safety distances from the release, 22 23 based on this threshold concentration limit, are determined and reported for each experiment 24 conducted.

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26 *Keyword:* CO₂ release, Under-expanded jet, Dispersion, Large-scale pipeline.

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1 1 Introduction

2 The importance of anthropogenic CO₂ in the atmosphere driving rapid global warming (https://www.iea.org/topics/climatechange/) and the recognition of the need to reduce CO₂ 3 4 emissions are now widely accepted. This has motivated research into various emissions mitigation/reduction technologies, the most important of which is Carbon Capture and 5 6 Storage (CCS) [1]. CCS has the greatest potential to reduce atmospheric emissions of CO₂, mitigating some negative effects of the continued use of fossil fuels until clean energy 7 8 technologies can be implemented [2]. Its rapid deployment has the potential to contribute 9 significantly to limiting the rise in global temperatures to 2 °C above pre-industrial levels [3]. 10 Pipeline transportation of CO₂ for CCS is accepted as the safest and most efficient 11 transportation option [4]. However CO₂ pipelines present a significantly different risk profile compared to, for example, hydrocarbon pipelines. Notably CO2 is denser than air, odorless, 12 colorless and not flammable. The acute health effects of CO2 are dependent on the 13 concentration in the air and duration of exposure, concentrations of 10 % v/v will render an 14 15 adult unconscious after 1 minute and exposure to concentrations of 20 % v/v or greater is instantaneously fatal [5,6,7]. A threshold value of 5 % v/v concentration is assumed to be the 16 lower limit for adverse human effects [27]. Safety distances in published risk assessments for 17 18 underground high pressure CO_2 pipelines vary from less than 1 m to 7.2 km as a result [8]. This large variation in the safety distance has significant implications for pipeline design, 19 routing, operation, maintenance and security. 20

21 In the event of a rupture in a high pressure CO₂ pipeline a significant mass of inventory may

be discharged very rapidly, possibly before the leak is detected. Due to its relatively high 1 Joule Thomson expansion coefficient, CO₂ may reach temperatures as low as -70 °C during a 2 3 pipeline release. This has the potential to induce brittle fracture in a pipe wall in addition to 4 the initial risk of ductile fracture [9], as well as introducing the possibility of dry ice 5 formation in the pipeline and near-field dispersion zone [10]. During a prolonged release a 6 fully expanded dispersion cloud will be connected to the pipeline rupture by a highly 7 under-expanded jet containing a Mach disk orthogonal to the flow direction [11]. External to the pipeline, solid CO₂ particles are most likely to form in the jet. If solid CO₂ is formed it 8 9 will subsequently sublime and mix with the vapor cloud. Transient cloud properties may be 10 significantly affected by the formation or absence of solid CO₂ in the under-expanded jet. In the process of mixing with the atmosphere the momentum of the jet diminishes and 11 12 dispersion will continue as a vapor cloud dispersion [12].

Many experimental studies have been conducted to establish a clear understanding of the 13 hazards associated with the failure of CO₂ pipelines. As part of the CO₂PipeHaz project, 14 INERIS [13-15] built a 2 m³ spherical vessel connected to a 9 m long pipe with an inner 15 16 diameter of 50 mm. This apparatus was used to measure temperatures and gas concentrations in the dispersion region during outflow from the pipe in order to guide large scale 17 18 experimental CO₂ release studies. An important observation from this work was that 19 significant solids are generated within the near-field of dense phase releases, despite the release itself containing no dry ice. Witlox et al. [23,24] presented experimental work 20 conducted by BP and Shell during the CO₂ PIPETRANS JIP, including both high-pressure 21

steady-state and time-varying cold CO₂ releases and high-pressure time-varying supercritical 1 hot CO₂ releases. For all cases solid CO₂ formed in the dispersion area and was observed to 2 sublime rapidly and no rainout was predicted. Xie et al. [16,17] developed a 23 m long 3 4 circulating pipeline with a 30 mm inner diameter to study the pipeline leakage process of 5 supercritical CO₂ in a vertical direction. A typical highly under-expanded jet flow structure 6 was observed. However, this structure disappeared as the orifice size increased. DNV-GL [18] investigated the discharge of liquid CO_2 from a 0.5 m³ pressurized vessel equipped with an 7 8 actuator valve. The results showed that the CO₂ concentrations near the orifice depend mainly 9 on the jet shape rather than the mass flow rate. The concentrations at 9 and 15 m from the 10 release point tended to increase continuously while saturated liquid was being discharged, and then to drop with the transition to vapor outflow. Xing et al [19] used a series of scaling 11 12 rules based on field experiments of CO₂ dispersion to simulate large scale CO₂ blowouts. Through comparison with the $k-\epsilon$ model and statistical performance indicators, it was 13 concluded that the scaling rules appeared applicable to field experiments of accidental 14 release. 15

Several experimental research programs have been performed at the Spadeadam Test Site in Cumbria, UK. The COSHER JIP [20] performed a large scale pipeline rupture test using a 226.6 m long pipeline loop built with 219.1 mm diameter steel pipe and fed from both ends by a 148 m³ reservoir of CO₂. The results showed that a visible cloud reached a maximum height of about 60 m and a maximum distance from the rupture location of about 400 m. Pseudo-steady CO₂ concentrations were reached at up- and downwind locations in the near-field in low wind speed conditions, but not in the far-field. Wareing et al. [21,22] studied
the venting of dense and gas phase CO₂ through a single, straight vertical vent pipe within the
framework of the COOLTRANS research program. The experimental data used in these
releases was used to develop a CO₂ dispersion model.

As part of the CO₂QUEST project [25,26], this paper reports work to study highly under-expanded jets and the dispersion characteristics of gaseous and dense phase CO₂ during sudden release from a pipeline. Six large-scale CO₂ release experiments are reported. The pipeline used was 258 m long with an inner diameter of 233 mm, three orifice sizes were used in experiments (15 mm, 50 mm and Full Bore Rupture). The experimental studies provide a detailed understanding of the hazards presented by CO₂ releases, the data recorded could be used to validate outflow and dispersion models.

12 **2** Experiments

13 2.1 Experimental setup

14 Figure 1 shows a schematic representation of the CO_2 pipeline employed for conducting the 15 release experiments. The pipeline was 257 m long with and inner diameter of 233 mm and a 16 wall thickness of 20 mm. It was built with 16MnR low temperature carbon steel and had a 17 maximum pressure rating of 16 MPa. Concrete foundations and supports rigidly clamped the pipeline 1.3 m above the ground. Additionally, a reinforcing anchor device capable of 18 19 resisting an acting force of 400 kN was designed to prevent movement of the pipeline during experiments. The pipeline was wrapped in 50 kW heating tape and a 50 mm thick layer of 20 21 thermal insulation. The heating system could warm the inventory to a maximum temperature

1 of 40 °C.

2	A 1 m long dual-disc blasting pipe built using grade 304 stainless steel was attached to one
3	end of the pipeline and used to initiate experiments. The blasting device consisted of two
4	rupture discs and two disc holders, a solenoid valve, two pipe sections and an end flange.
5	Replaceable flanges with different sizes of pre-prepared (circular) orifices were installed in
6	the blasting pipe for experiments, orifice diameters of 15 mm, 50 mm and Full Bore Rupture
7	(FBR) were used. Each orifice was cut in the center of the flange. To initiate release
8	experiments disk A was ruptured by reducing the pressure in section 1 of the blasting pipe
9	(see Figure 1). This increased the pressure differential across disk B causing it to rupture, the
10	net result is the essentially instantaneous opening of the pipeline.
11	The pipeline was charged and experiments conducted as follows:
12	(1) Purge the pipeline using gaseous CO ₂ .
13	(2) Charge the pipeline with the previously calculated mass of CO_2 required for the
14	experiment.
15	(3) Once charged, use the heating system to alter inventory conditions to those desired for
16	the experiment.
17	(4) Isolate the experimental field.
18	(5) Initiate the experiment using the dual-disc blasting device and record the desired
19	experimental data.
20	(6) After the release is complete and instruments indicate the cloud is fully dispersed
21	prepare the experimental field for the next release.

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2 2.2 Pipeline instrumentation

An overhead schematic view of the locations and types of instruments in the dispersion zone is shown in Figure 2. Thermocouples and CO₂ concentration sensors were both arranged on vertical tubes at the same height as the pipeline, as shown in Figure 3. T-type thermocouples with an uncertainty of ±1 °C, a measurement range of -200 °Cto 400 °C and response times of 100 ms were used. The CO₂ concentration sensors used were COZIR-W type manufactured by Gas Sensing Solutions Ltd (GSS). Their measurement range was 0–100 %, their accuracy was ±3 % and their response time was 4 s.

10 Two data acquisition systems ran simultaneously during experiments, an NI cRIO-9025 11 system which was used to sample the thermocouples while an RS485 communication system 12 was used to sample the CO₂ concentration sensors. The NI cRIO-9025 system consisted of 13 one 9025 and one 9144 chassis and two NI 9213 thermocouple input modules. The RS485 14 communication bus adopted a twist-pair with RVVSP 2×2×0.5 mm² and a master-slave half 15 duplex mode. The data-acquisition code was programmed using LabVIEW software from 16 National Instruments UK.

A weather station was established to record ambient temperature, pressure, humidity, wind speed and direction. The ambient pressure was measured using a QA-1 air pressure sensor with an accuracy of ± 0.03 kPa and a range of 55 kPa to 106 kPa. The ambient temperature and humidity were measured using PTS-S environment monitoring sensors, each with an uncertainty of ± 0.1 °C and ± 0.2 % and a range of -50 °C to 80 °C and 0 to 100 % respectively. The wind speed and direction were measured using an EC-A1 ultrasonic wind sensor with an
 accuracy of ±0.01 m/s and ±1° and a range of 0 to 60 m/s and 0 to 360°. Several digital HD
 video cameras and a Phantom 2 Vision aerial drone were used to record the evolution of the
 visible cloud during experiments.

5 2.3 Experiments conducted

6 Six CO_2 release experiments were performed to investigate dispersion behaviour during the 7 release of gaseous and dense phase CO_2 from a pipeline. Orifice diameters of 15 mm, 50 mm 8 and Full Bore Rupture were used. The purity of the CO_2 was 99.9%. The initial and 9 environmental conditions of six tests are presented in Table 1. For tests 1 and 4 the ambient 10 pressure, temperature, wind speed and direction fluctuated violently as result of unstable 11 atmospheric conditions during the long depressurization. Other tests were carried out in more 12 stable ambient conditions.

13 **3** Experimental results

14 3.1 Gas phase tests

15 3.1.1 Visible cloud development

Based on the experimental data, the development of the visible cloud may be divided into
three stages: (I) rapid expansion, (II) the metastable stage and (III) the slow attenuation stage.
For tests 1, 2 and 3 the duration times for the respective clouds in each stage of development
are shown in Table 2 together with the total depressurization times.

Figures 4, 5 and 6 show the development of the visible cloud for tests 1, 2 and 3 respectively.

As may be observed in each figure the released gas may be seen as a white cloud with a base

1 touching the rupture plane. Although the inventory is colourless, the significant cooling of the CO₂ as it expanded passing through the orifice and in the under-expanded jet resulted in the 2 3 formation of solid CO₂ particles. These particles remained entrained in the cloud and did not 4 rain out. For tests 1 and 2, the visible cloud was fully developed during the rapid expansion 5 phase and its dimensions remained relatively stable during the metastable stage. During the 6 attenuation stage the expansion angle of the jet and overall dimensions of the cloud 7 decreased. The white visible cloud disappeared in the forefront of the jet flow, indicating that 8 the dry ice partcles had gradually sublimated during diffusion.

9 For test 2 the jet travelled in a straight line along the axial direction throughout the 10 experiment, the divergent angle remained circa 11° throughout. In the metastable stage the 11 visible cloud dimensions remained relatively constant with a length of circa 10 m and a 12 maximum radius of circa 1.5 m. As may be observed at 45 s and 60 s (figure 5), the 13 intercepting shock and the Mach disc were clearly visible in the jet flow.

For test 3 jet velocities are indicated in figure 6 for the first 0.8 s. In the rapid expansion stage the visible white cloud rapidly expanded, entraining dry ice particles and condensed water vapour. The expanding jet velocity simultaneously decreased as the distance from the rupture increased. The visible cloud reached a maximum length of circa 40 m. The divergent angle of the jet was circa 18° . In the slow attenuation stage the visible white cloud and the mixture of gaseous CO₂, air and raised dust separated. Even when no white cloud was visible, a measurable CO₂ cloud continued to spread, relying on momentum, wind and body forces to do so. As test 3 involved an FBR the dry ice particles formed were spread farther, allowing
 the considerable diffusion velocity and range of this test to be observed.

3 3.1.2 Temperature distribution

Figure 7 shows the temperature evolution along the axial line of the discharge area in tests 1, 4 5 2 and 3. In all cases, after rupture the ambient temperature in the far-field dropped as a result 6 of the expansion of the escaping gas and the sublimation of dry ice particles. The cloud 7 temperature along the axis of the release gradually rose as the distance from the orifice 8 increased due to the reduction of the cloud velocity and the fraction of dry ice further from 9 the orifice. When the release was complete the temperature of the CO₂ gradually increased to 10 ambient as it continued to mix with air. For test 1, the temperature distribution in the dispersion region was small but it fluctuated as the smaller jet in this experiment was more 11 12 significantly affected by the unstable atmospheric environment. For test 2, the temperature contour lines quickly extended along the axial direction and reached the maximum values at 13 14 15 s after rupture. For test 3 the rates of extension and retreat of the temperature contour lines were similar as a result of the violent mixing of the escaping gas and air and the extremely 15 16 unstable atmosphere in the full bore release. The maximum temperature drops observed 4 m from the orifice were 4.2 °C, 5.5 °C and 17.6 °C, occuring at the times of 35 s, 15 s and 7 s 17 for tests 1, 2 and 3 respectively. The lowest cloud temperatures were only observed for short 18 19 times, while the maximum temperature drop ampitudes became bigger with increasing orifice 20 diameter due to the more violent mixing of the escaping gas and air.

Figure 8 shows the lowest temperature distribution in the discharge area for tests 1, 2 and 3. 1 The y direction is along the jet axis and the x direction is horizontal distance. Obviously, the 2 3 temperature of the escaping gas increased along the x and y directions in the discharge area. 4 The lengths of the temperature contour lines along the x direction were much longer than 5 those along the y direction while the temperature gradients along the x direction were much lower. For the 23 °C and 24 °C contours recorded for test 1, the 23 °C, 24 °C and 25 °C 6 contours recorded for test 2, and the 10 °C, 13 °C and 16 °C contours recorded for test 3, 7 the length ratios between the y and x directions were 16.1, 12.9, 33.5, 29.6, 21.3, 58.3, 30.4 8 9 and 24.1 for each contour line respectively. This confirms that the temperature contour lines along the y direction were much larger than that along the x direction as a result of the much 10 greater axial velocity of CO₂ compared to radial velocity. The length ratio between the 11 12 temperature contour lines along the x and y directions became larger with increasing orifice diameter. This was mainly because the jet velocities and distribution range became greater 13 with the increase in orifice diameter. For tests 1, 2 and 3, the length and width of the 14 low-temperature zones were predicted to be 13 by 2.5 m, 16 by 2.2 m, and 20 by 2.5 m 15 16 respectively.

17 3.1.3 CO₂ concentration dispersion

Figure 9 shows the evolution of concentration along the axial direction in tests 1, 2 and 3. As may be observed, the contours of CO_2 concentration remained at their maximum extent through the metastable state. The closer the distance to the orifice, the longer the exposure to 5 % v/v concentrations of CO_2 . The start time of the contour of low CO_2 concentration was earlier than that of high CO₂ concentration, this was due to the response lag of the CO₂
 concentration sensors to the high-velocity gas. The response lag had little influence on the
 assessment of dangerous concentration distances.

For test 1, the 5 % CO₂ concentration contour reached a maximum distance of 9.2 m from the 4 5 release orifice, thus the safety distance along the axial direction should be less than 10 m in 6 test 1. For test 2, at the end of the release (159 s), CO₂ concentrations within the measurement 7 area stayed above 3 % concentration, decreasing below 1 % concentration by 207 s after the 8 rupture. Thus for test 2 the safety distance along the axial direction should be at least 12 m. 9 For test 3, the 5 % CO₂ concentration contour quickly reached its maximum extent at 13 s. CO₂ concentrations at a distance of 5 m from the release orifice reached 30.1 % at the end of 10 the release (15 s) and reached a maximum value at 27 s. At this distance the CO_2 11 12 concentration remained above 5 % v/v until 150 s after rupture. This persistent concentration was caused by the sublimation of the dry ice particles in the discharge area. The safety 13 14 distance in test 3 should be at least 25 m. It was obvious that the larger discharge diameter 15 brought a greater amount of discharged CO₂ into the dispersion region over a given time 16 interval.

17 3.2 Dense phase tests

18 3.2.1 Visible cloud development

For tests 4, 5 and 6 the duration times for the respective clouds in each stage of developmentare shown in Table 3 together with the total depressurization times. Compared with the gas

phase tests, the duration times of the clouds in each stage of evolution were longer, as
 expected.

3 Figures 10, 11 and 12 show the development of the visible clouds for tests 4, 5 and 6 respectively. The recorded dimensions of the test 4 dispersion cloud at various times are 4 5 given in table 4. The data reports a gradual increase in the length and height of the visible 6 cloud as the expanding velocity decreased from an initial peak. For tests 5 and 6 cloud 7 dimensions are also reported in table 4. The data indicates that the size and expanding velocity of the visible cloud increased as the orifice size increased. For test 4, the dimensions 8 9 of the visible cloud reached their maximum at 9 s. For tests 5 and 6, the clouds reached their 10 maximum horizontal dimensions at 6 s and 5 s respectively, although maximum visible cloud heights were achieved earlier (see table 4). The dispersion clouds therfore continued to 11 12 expand horizontally after reaching their maximum visible height, solid CO₂ formation and gas density contributed to this behaviour. Orifice size had a clear influence on visible cloud 13 height; the angle of reflection of the jet from the ground was greater as the orifice size 14 15 increased, resulting in higher dispersion clouds.

For the dense phase tests 4, 5 and 6, the divergent angles of the jets were greatest at the moment of rupture and decreased gradually during blowdown. In the rapid expansion stage the axial length of the jet increased quickly and the leading edge of the jet expanded along an arc frontage. In the metastable stage the dimensions of the cloud remained essentially stable and the forward edge of the cloud adopted a fan-shape. The heavy gas effect started prompting the free diffusion of CO_2 in low-lying areas. In the slow attenuation stage, as the 1 expanding jet velocity decreased cloud diffusion increasingly depended on residual 2 momentum. The size of the visible cloud began to decay and the attenuation velocity 3 decreased. Compared to the gas phase tests the visible cloud produced in the dense phase 4 tests had a much wider dispersion range and a greater amount of dry ice particles and 5 condensed water vapour were formed. The amount of condensed water vapour entrained in 6 the jet was dependent on the ambient humidity and the degree of cooling of the CO_2 in the jet 7 (and therefore on the initial inventory pressure).

8 3.2.2 Temperature distribution

9 Figure 13 shows the temperature evolution along the axial line of the discharge area in tests 5 10 and 6. For test 4 the temperatures in the dispersion area were not recorded due to instrument failure. For test 5, the temperature contour lines quickly extended along the axial direction 11 12 and reached their maximum extent at 13 s. The temperature drop amplitude 5 m from the rupture reached 31 °C, a larger drop than in test 2. For test 6, the temperature drop amplitude 13 14 reached 24 °C at 3 s, a larger drop than that at the same time for test 3, and reached a maximum value of 102 °C at 17 s. This time was later than the equivalent time in test 3 (3 s) 15 16 due to the continuous sublimation of dry ice particles in test 6. The results indicate similar behaviour in the temperature evolution along the release direction between the dense and gas 17 phase tests. However, it is clear that for a given orifice size the low-temperature areas in the 18 19 dense phase tests covered a greater area than in the gas phase tests.

Figure 14 shows the distribution of low temperature zones in the discharge area for tests 5and 6. The y direction is along the jet axis and x direction is the horizontal distance. The

overall trend was for the temperature gradient along the x direction to be much more abrupt 1 than along the y direction, while the lengths of the temperature contour lines along the x 2 direction were much longer than those along the y direction. For 0 °C, 3 °C, 6 °C and 9 °C in 3 test 5, the length ratios between the temperature contour lines along the y direction and the x 4 5 direction were close to 10.1. This indicated that the temperature distribution shifted to the right due to the effect of the southeast wind. For the -40 °C, -30 °C, -20 °C and -10 °C 6 7 contour lines in test 6, the length ratios between the y and x directions were 96.6, 45.6, 32.7 8 and 27.1 respectively. This shows that the diffusion velocity in the FBR release with dense 9 phase CO₂ was extremely fast, as a result the temperature distribution in the resulting 10 dispersion cloud could be considered essentially axisymmetric. According to the contour extending trend in tests 5 and 6, it was predicted that the maximum length and width of the 11 12 low-temperature zone was 16 m by 3 m and 30 m by 5 m in tests 5 and 6 respectively.

13 3.2.3 CO₂ concentration dispersion

Figure 15 shows the evolution of CO₂ concentration along the axis of release in tests 4, 5 and 14 6. For test 4, CO₂ concentration 5 m from the release orifice reached 5 % v/v 15 s after 15 16 rupture and remained at this quasi steady state level until 3200 s. The greatest extent of the 5 % concentration contour was 19.2 m from the orifice, reached 263 s after rupture. Therefore 17 the safety distance along the release direction should be circa 20 m for test 4. For test 5, the 18 19 concentration contour lines fluctuated intermittently due to the varying southeast wind. The 20 5 % v/v concentration contour extended 12 m from the release orifice by 6 s and remained above 5 % at this distance until the end of the release (482 s). The safety distance along the 21

release direction in test 5 should be circa 60 m. For test 6, CO₂ concentration contours began 1 2 to extend from the rupture after 6 s and reached their farthest point at 15 s. At the end of the 3 release (40 s) CO₂ concentrations at a distance of 60 m from the release orifice had remained above 5 % v/v concentration as a result of the continual sublimation of the dry ice particles. 4 5 61 % v/v concentration was achieved 20 m from the orifice (the maximum at this location) 6 earlier than a 100 % v/v concentration was achieved 5 m from the orifice. This suggested that 7 due to dry ice sublimation and the response time of the sensors, the duration of exposure to maximum CO₂ concentrations was longer as the distance to the orifice decreased. The safety 8 9 distance along the release direction in test 6 reached circa 160 m.

10 4 Discussion

This paper studied highly under-expanded jets and their dispersion characteristics during the 11 12 release of gaseous and dense phase CO₂ from a large-scale pipeline. Such a large capacity pipeline was essential as it permitted long duration experiments to be performed and large 13 14 amounts of data to be captured for analysis. A great many sensors were used to monitor the formation of the visible cloud and the variation of temperatures and concentrations in the 15 16 far-field. The research results of near-field source terms and dispersion behavior in this study are necessary and of paramount importance for assessing safety distances and the impact of 17 CO₂ pipeline releases on the surrounding environment. 18

19 Rupture or puncture of a high pressure CO_2 pipeline will almost certainly result in a 20 high-velocity jet from the pipeline and the transition of the inventory through different 21 physical states. For high pressure pipelines, sonic velocity will likely be reached at the outlet

of the pipe and the resulting free jet will therefore be sonic. This would lead to a highly under 1 expanded flow that contains a Mach disk, the precise form of which depends on the ratio of 2 3 the exit to atmospheric pressure. For CO₂ releases, the temperature and pressure decrease that 4 accompanies inventory expansion will lead to the formation of dry ice particles and 5 condensed water vapor. Likewise, sublimation of dry ice particles will result in heat removal from the gas phase and an associated temperature decrease. Formation of solid CO₂ will 6 7 affect the shape and properties of a CO_2 cloud in ways different to a gas/liquid cloud [28]. In addition, ground topography and physical objects, as well as wind direction, may have a 8 9 significant influence on the spread and movement of a CO₂ cloud. These extremely 10 complicated phenomena may be expected during the dispersion process of gaseous or dense phase CO₂ during sudden release. High pressure CO₂ dispersion modelling therefore requires 11 12 appropriate source terms, accurate turbulence modeling, a three-phase accurate equation of state, to account for heat transfer between the ground and the flowing fluid and precise 13 14 particle tracking techniques for estimating the amount of solid CO₂ [29-31]. No mathematical model is currently capable of predicting all these phenomena during complex releases. 15

16 Currently, the authors focus on experimental research over theoretical analysis. This and 17 related large-scale experimental work contributes basic data to CO_2 dispersion research and 18 can be used to validate outflow, near-field and far field dispersion models. Experimental 19 characterisation of the temperatures in the immediate vicinity of a release will also be 20 performed to enable estimation of the risk to piping, plant or structures from low temperature 21 embrittlement. For the design, construction and operation of new high pressure CO_2 pipelines through populated areas, the safety distance to the release point, measured along the release axis, should be consistent with the 5 % v/v concentration threshold in the dispersion cloud. This distance is critically important when performing consequence failure analysis and quantifying major hazards for CO₂ pipelines. More sophisticated experimental and theoretical studies will be conducted on dispersion behavior during sudden releases in the future.

6 5 Conclusions

7 This article has presented the results of a large-scale experimental study of under-expanded
8 jets and dispersion characteristics of gaseous and dense phase CO₂ following pipeline rupture
9 through three orifice sizes (15 mm, 50 mm and Full Bore Rupture). The following
10 conclusions were drawn from the study:

(1) A highly under-expanded flow containing a Mach disk was developed during the release
of high pressure CO₂. A large quantity of dry ice particles formed in the near-field due to
Joule-Thomson cooling, these were carried into the far-field. The dispersing cloud was made
visible by both these dry ice particles and condensing water vapor. The dry ice sublimed
rapidly and did not settle.

16 (2) After rupture, the ambient temperature in the far-field dropped as a result of the expansion 17 of the escaping gas and the sublimation of the dry ice particles. The cloud temperature along 18 the release axis gradually rose as the expanding velocity and the fraction of dry ice decreased 19 along this axis. For given initial conditions, as the orifice diameter increased the degree of 20 cooling and the dispersion distance of CO_2 in the discharge area, and therefore the safety 21 distance, increased.

1	(3) Compared to the gas phase tests, the visible cloud produced in dense phase tests entrained
2	more condensed water vapour, included a greater mass of dry ice and had a much wider
3	dispersion range. The footprint of the low-temperature areas of the cloud and the safety
4	distances in the dense phase tests were also much greater than that in the gas phase tests. The
5	dense CO ₂ gas was observed to gather in low lying areas after release when not dispersed by
6	the wind.

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12		
13		
14		References
15	[1]	Haszeldine RS. Carbon capture and storage: how green can black be? Nature 2009;
16		325:1647-1652.
17	[2]	Munkejord ST, Hammer M, Løvseth SW. Intergovernmental panel on climate change,
18		carbon capture & storage, ISBN 92-9169-119-4. Appl Energy 2016; 169:499-523.
19	[3]	IEA, Energy Technology Perspectives 2008, Paris: France, 2008.
20	[4]	Chong FK, Lawrence KK, Lim PP, Poon MCY, Foo DCY, Lam HL, Tan RR PP,

21 Planning of carbon capture storage deployment using process graph approach. Energy

1 2014; 76:641-651.

2	[5]	Iribarren D, Petrakopoulou F, Dufour J. Environmental and thermodynamic evaluation
3		of CO2 capture, transport and storage with and without enhanced resource recovery.
4		Energy 2013; 50:477-85.
5	[6]	Duncan IJ, Wang H. Estimating the likelihood of pipeline failure in CO ₂ transmission
6		pipelines: New insights on risks of carbon capture and storage. Int J Greenh Gas Con
7		2014; 21: 49-60.
8	[7]	Brown S, Beck J, Mahgerefteh H. Global sensitivity analysis of the impact of impurities
9		on CO ₂ pipeline failure. Reliab Eng Syst Safe. 2013; 115: 43–54.
10	[8]	Molag M, Dam C. Modelling of accidental releases from a high pressure CO ₂ pipelines.
11		Energy Procedia 2011; 4:2301-2307.
12	[9]	Botros KK, Hippert E Jr, Craidy P. Measuring decompression wave speed in CO ₂
13		mixtures by a shock tube. Pipelines International 2013; 16:22-28.
14	[10]	Mazzoldi A, Hill T, Colls JJ. CO ₂ transportation for carbon capture and storage:
15		Sublimation of carbon dioxide from a dry ice bank. Int J Greenh Gas Con 2008
16		2:210-218.
17	[11]	Witlox HWM, Harper M, Oke A. Modelling of discharge and atmospheric dispersion for
18		carbon dioxide releases. J Loss Prevent Proc 2009; 22:795-802.
19	[12]	Witkowski A, Rusin A, Majkut M, Rulik S, Stolecka K. Comprehensive analysis of
20		pipeline transportation systems for CO ₂ sequestration. Energ Convers Manage 2013; 76:
21		665-673.

1	[13] Woolley RM, Fairweather M, Wareing CJ, Falle SAEG, Proust C, Hebrard J, Jamios D.
2	Experimental measurement and Reynolds-averaged Navier-Stokes modelling of the
3	near-field structure of multi-phase CO ₂ jet releases. Int J Greenh Gas Con 2013; 18:139–
4	149.
5	[14] Gant SE, Narasimhamurthy VD, Skjold T, Jamois D, Proust C. Evaluation of
6	multi-phase atmospheric dispersion models for application to Carbon Capture and
7	Storage. J Loss Prevent Proc 2014; 32:286-298.
8	[15] Woolley RM, Fairweather M, Wareing CJ, Falle SAEG, Mahgerefteh H, Martynov S,
9	Brown S, Narasimhamurthy VD, Storvik IE, Sælen L, Skjold T, Economou IG,
10	Tsangaris DM, Boulougouris GC, Diamantonis NI, Cusco L, Wardman M, Gant SE,
11	Wilday J, Zhang YC, Chen SY, Proust C, Hebrard J and Jamois D. CO ₂ PipeHaz:
12	quantitative hazard assessment for next generation CO ₂ pipelines. Energy Procedia 2014,
13	63:2510–2529.
14	[16] Xie QY, Tu R, Jiang X, Li K, Zhou XJ. The leakage behavior of supercritical CO ₂ flow
15	in an experimental pipeline system. Appl Energy 2014; 130:574-580.
16	[17] Li K, Zhou XJ, Tu R, Xie QY, Jiang X. The flow and heat transfer characteristics of
17	supercritical CO ₂ leakage from a pipeline. Energy 2014; 71:665-672.
18	[18] Ahmad M, Osch MB, Buit L, Florisson O, Hulsbosch-Dam C, Spruijt M, Dacolio F.
19	Study of the thermohydraulics of CO ₂ discharge from a high pressure reservoir. Int J
20	Greenh Gas Con 2013; 19:63-73.
21	[19] Xing J, Liu ZY, Huang P, Feng CG, Zhou Y, Sun RY, Wang SG. CFD validation of

1	scaling rules for reduced-scale field releases of carbon dioxide. Appl Energy 2014; 115:
2	525-530.
3	[20] Ahmad M, Lowesmith B, Koeijer Gd, Nilsen S, Tonda H, Spinelli C, Cooper R, Clausen
4	S, Mendes R, Florisson O. COSHER joint industry project: Large scale pipeline rupture
5	tests to study CO ₂ release and dispersion. Int J Greenh Gas Con 2015; 37:340–353.
6	[21] Wareing CJ, Fairweather M, Falle SAEG, Woolley RM. Validation of a model of gas and
7	dense phase CO ₂ jet releases for carbon capture and storage application. Int J Greenh
8	Gas Con 2014; 20:254-271.
9	[22] Wen J, Heidari A, Xu BP, Jie HG. Dispersion of carbon dioxide from vertical vent and
10	horizontal releases-A numerical study. J Process Mechanical Engineering 2013;
11	227:125-139.
12	[23] Witlox HWM, Harper M, Oke A, Stene J. Validation of discharge and atmospheric
13	dispersion for unpressurised and pressurised carbon dioxide releases. Process Saf
14	Environ 2014; 92:3-16.
15	[24] Witlox HWM, Harper M, Oke A, Stene J. Phast validation of discharge and atmospheric
16	dispersion for pressurised carbon dioxide releases. J Loss Prevent Proc 2014;
17	30:243-255.
18	[25] Brown S, Martynov S, Mahgerefteh H, Fairweather M, Woolley RM, Wareing CJ, Falle
19	SAEG, Rutters H, Niemi A, Zhang YC, Chen SY, Besnebat J, Shah N, Dowell NM,
20	Proust C, Farret R, Economou IG, Tsangaris DT, Boulougouris GC, Wittenberghe JV.
21	CO ₂ QUEST: Techno-economic assessment of CO ₂ quality effect on its storage and

transport. Energy Procedia 2014; 63:2622-2629.

2	[26] Guo XL, Yan XQ, Yu JL, Zhang YC, Chen SY, Mahgerefteh H, Martynov S, Collard A,
3	Proust C. Pressure response and phase transition in supercritical CO2 releases from a
4	large-scale pipeline. Appl Energy 2016; 178:189-197.
5	[27] Kruse H, Tekiela M. Calculating the consequences of a CO ₂ -pipeline rupture. Energy
6	Convers 1996; 37:1013-1018.
7	[28] Harper P, Wilday J, Bilio M. Assessment of the Major Hazard Potential of Carbon
8	Dioxide (CO ₂). Health and Safety Executive 2011; 1-28.
9	[29] Zhou XJ, Li K, Tu R, Yi JX, Xie QY, Jiang X. A modelling study of the multiphase
10	leakage flow from pressurized CO ₂ pipeline. J Hazard Mater 2016; 306:286-294.
11	[30] Wareing CJ, Woolley RM, Fairweather M, Falle SAEG. A composite equation of state
12	for the modeling of soniccarbon dioxide jets in carbon capture and storage scenarios.
13	AIChE J 2013; 59:3928-3942.
14	[31] Wareing CJ, Fairweather M, Falle SAEG, Woolley RM. Modelling punctures of buried
15	high-pressure dense phase CO ₂ pipelines in CCS applications. Int J Greenh Gas Con
16	2014; 29:231-247.
17	



Fig. 1 Process flow.



Fig. 2 Distribution of measurement points in discharge area.



Fig. 3 CO2 concentration sensor and installation.



Fig. 4 Visible cloud development of the gaseous CO₂ release experiments with 15 mm orifice.



Fig. 5 Visible cloud development of the gaseous CO_2 release experiments with 50 mm orifice.



Fig. 6 Visible cloud development of the gaseous CO₂ release experiments with the full bore orifice.



Fig. 7 Temperature evolutions along the centerline of the gaseous CO_2 release experiments with three different orifices (15 mm, 50 mm and FBR).



Fig. 8 Temperature distribution area of the gaseous CO_2 release experiments with three different orifices (15 mm, 50 mm and FBR).



Fig. 9 CO₂ concentration development along the centerline of the gaseous CO₂ release experiments with three different orifices (15 mm, 50 mm and FBR).



Fig. 10 Visible cloud development of the dense CO_2 release experiments with 15 mm orifice.



Fig. 11 Visible cloud development of the dense CO_2 release experiments with 50 mm orifice.



Fig. 12 Visible cloud development of the dense CO₂ release experiments with the full bore orifice.



Fig. 13 Temperature evolutions along the centerline of the dense CO₂ release experiments with three different orifices (15 mm, 50 mm and FBR).



Fig. 14 Temperature distribution area of the dense CO_2 release experiments with three different orifices (15 mm, 50 mm and FBR).



Fig. 15 CO₂ concentration development along the centerline of the dense CO₂ release experiments with three different orifices (15 mm, 50 mm and FBR).