# Dust, CO, and [C1]: cross-calibration of molecular gas mass tracers in metal-rich galaxies across cosmic time

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Accepted 2022 July 25. Received 2022 July 25; in original form 2022 February 13

## ABSTRACT

We present a self-consistent cross-calibration of the three main molecular gas mass tracers in galaxies, namely the  ${}^{12}CO(1-0)$ ,  $[C1](^{3}P_{1}-^{3}P_{0})$  lines, and the submm dust continuum emission, using a sample of 407 galaxies, ranging from local discs to submillimetre-selected galaxies (SMGs) up to  $z \approx 6$ . A Bayesian statistical method is used to produce galaxy-scale universal calibrations of these molecular gas indicators, that hold over 3–4 orders of magnitude in infrared luminosity,  $L_{IR}$ . Regarding the dust continuum, we use a mass-weighted dust temperature,  $T_{\rm mw}$ , determined using new empirical relations between temperature and luminosity. We find the average L/ $M_{mol}$  gas mass conversion factors (including He) to be  $\alpha_{850} = 6.9 \times 10^{12} \text{ W Hz}^{-1} \text{ M}_{\odot}^{-1}$ ,  $\alpha_{\rm CO} = 4.0 \,\mathrm{M}_{\odot} \,(\kappa \,\mathrm{km \, s^{-1} \, pc^2})^{-1}$ , and  $\alpha_{\rm CI} = 17.0 \,\mathrm{M}_{\odot} \,(\kappa \,\mathrm{km \, s^{-1} \, pc^2})^{-1}$ , based on the assumption that the mean dust properties of the sample ( $\kappa_{\rm H}$  = gas-to-dust ratio/dust emissivity) will be similar to those of local metal rich galaxies and the Milky Way. The tracer with the least intrinsic scatter is [C1](1-0), while CO(1-0) has the highest. The conversion factors show a weak but significant correlation with  $L_{\rm IR}$  which is not apparent when  $T_{\rm mw}$  is held constant. Assuming dust properties typical of metal-rich galaxies, we infer a neutral carbon abundance  $X_{CI} = [C^0/H_2] = 1.6 \times 10^{-5}$ , similar to that in the Milky Way. We find no evidence for bi-modality of  $\alpha_{CO}$  between main-sequence (MS) galaxies and those with extreme star-formation intensity, i.e. ultraluminous infrared galaxies (ULIRGs) and SMGs. The means of the three conversion factors are found to be similar between MS galaxies and ULIRGs/SMGs, to within 10-20 per cent. The overarching conclusion of our work is that, for metal-rich galaxies, near-universal average values for  $\alpha_{CO}$ ,  $X_{CI}$ , and  $\kappa_{H}$  are adequate for global molecular gas estimates within the expected uncertainties. The 1 $\sigma$  scatter in our optimized values for  $\alpha_{\rm CO}$ ,  $X_{\rm CI}$ , and  $\kappa_{\rm H}$  are 0.14, 0.11, and 0.15 dex, respectively.

Key words: ISM: dust, extinction - Galaxies: high redshift - Submillimetre: galaxies, ISM - Radio lines: galaxies, ISM.

#### **1 INTRODUCTION**

The cosmic star formation rate (SFR) density has declined by more than an order of magnitude during the past  $\approx 8$  Gyr of cosmic history (Lilly et al. 1996; Madau et al. 1996; Madau & Dickinson 2014). The driver of star formation is the molecular gas supply in galaxies, and indeed the SFR–stellar mass (SFR– $M_{\star}$ ) relationship known as the galaxy main sequence (MS) is purely a by-product of the relationship between SFR and molecular gas (e.g. Baker et al. 2022), for unperturbed galaxies with significant gas reserves. A major observational goal is to produce a combined census of the molecular gas – the 'potential for future star formation' – and the stellar content – the 'record of past star formation' – over this period (e.g. Keres, Yun & Young 2003; Dunne et al. 2003, 2011; Zwaan et al. 2004; Zafar et al. 2013; Walter et al. 2014; Decarli et al. 2016, 2019; Saintonge et al. 2017; Driver et al. 2018; Rhee et al. 2018; Riechers et al. 2019).

The molecular gas fraction of a galaxy is a crucial component in models of galaxy formation (e.g. Obreschkow et al. 2009; Popping, Somerville & Trager 2014; Lagos et al. 2015; Chen et al. 2018) and thus measurements of  $H_2$  and stellar mass over large representative

galaxy samples are key requirements for understanding how galaxies have transformed from clouds of gas residing in dark matter haloes into the regular agglomerations of stars we see in the local Universe. While it is clear that CO(1–0)-luminous gas is the phase linked with star formation (e.g. Wong & Blitz 2002), observations of molecules with higher critical densities (e.g. HCN) revealed that it is the dense H<sub>2</sub> gas phase ( $n > 10^4$  cm<sup>-3</sup>) that correlates most tightly and linearly with tracers of star formation (Gao & Solomon 2004).

Atomic hydrogen (H I), on the other hand, constitutes a longer term gas reservoir for star formation, where under certain conditions of pressure, far-UV radiation field, density and metallicity, a phase transition H I  $\rightarrow$  H<sub>2</sub> takes place, catalysed by dust grains (e.g. Elmegreen 1993; Papadopoulos, Thi & Viti 2002; Blitz & Rosolowsky 2006): a picture supported by numerous observations (e.g. Honma, Sofue & Arimoto 1995; Leroy et al. 2008; Bigiel et al. 2008; Schruba et al. 2011). This transition occurs in the inner H I distribution of galaxies, in the cold neutral medium (CNM:  $n \sim 50-100 \text{ cm}^{-3}$ ,  $T_{kin} \sim 100-$ 200 K), meanwhile pure H I gas often extends many optical radii beyond the luminous stellar disc (e.g. Péroux & Howk 2020), where it can be found concomitant with cold dust (e.g. Thomas et al. 2002).

Unlike H<sub>I</sub> and its hyperfine line emission at 21 cm, the H<sub>2</sub> molecule in its S(0):J = 2 - 0 transition at 28 µm (the least

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excitation-demanding H<sub>2</sub> line) is essentially invisible at temperatures typical of giant molecular clouds (10–20 K). This is because its  $\Delta E/k_{\rm B} \sim 510 k$ , limits its excitation and detection only to shocked regions of molecular clouds, where gas temperatures can rise past  $\sim 1000$  K, for small ( $\sim 1-2$  per cent) gas mass fractions. Even then, to observe this H<sub>2</sub> line at 28 µm requires space-borne telescopes.

For these reasons, the rotational transitions of CO (the next most abundant molecule with  $[CO/H_2] \sim 10^{-4}$ ) are commonly used to trace H<sub>2</sub> gas, with the lowest transition (<sup>12</sup>CO J = 1-0) being the most established tracer. Its  $E_{10}/k_{\rm B} \sim 5.5$  K ensures a well-populated upper level even in the coldest gas, while its low critical density,  $n_{\rm cr} \sim 400$  cm<sup>-3</sup>, ensures its excitation even at low densities.<sup>1</sup>

The CO(1–0) line has significant optical depths in the typically macro-turbulent H<sub>2</sub> gas, though these arise locally within the velocity-coherent gas cells allowing the CO emission to trace gas mass throughout molecular clouds (e.g. Dickman, Snell & Schloerb 1986). The conversion factor,  $\alpha_{CO}$ , in the relation  $M_{H2} = \alpha_{CO}L'_{CO}$ cannot be determined using standard optically thin line formation physics due to the high line optical depths. This created the need for a  $\alpha_{CO}$  calibration as soon as the ubiquity of CO line emission in H<sub>2</sub> clouds was established. Observational and theoretical investigation of  $\alpha_{CO}$  suggests it is sensitive to metallicity, molecular gas surface density, and kinematic state in galaxies (e.g. Pelupessy & Papadopoulos 2009; Narayanan et al. 2011; Papadopoulos et al. 2012a; Bolatto, Wolfire & Leroy 2013).

Three distinct problems are now recognized regarding the use of CO as a global tracer of  $H_2$  mass in galaxies:

(i) The  $\alpha_{CO}$  factor is sensitive – in a highly non-linear fashion – to the ISM metallicity and ambient far-UV radiation fields (e.g. Israel 1997; Pak et al. 1998; Bolatto et al. 2013).

(ii) Non-self-gravitating molecular clouds – and/or very different average ISM states in terms of average temperature and gas density range from those found in spiral galaxies where  $\alpha_{CO}$  was first calibrated – can yield systematically different  $\alpha_{CO}$  factors. For example,  $\alpha_{CO} \sim 1/5 - 1/4 \times$  Galactic was initially reported for a sample of four ULIRGs by Downes & Solomon (1998).

(iii) Elevated cosmic ray (CR) energy densities can destroy CO below a certain gas density threshold, leaving behind more C-rich gas. This density threshold depends on the CR energy density in a highly non-linear fashion, as explored by Bisbas, Papadopoulos & Viti (2015), who found that regions of CO suppression may occur even in moderately enhanced CR conditions if the gas density is low, while the very high CR energy densities expected in ultraluminous infrared galaxies (ULIRGs) may be partly compensated by higher gas densities in such starbursts. Modelling [C I/CO] ratios as a function of CR, turbulence, gas density, and metallicity is an active area of theoretical research (e.g. Bisbas et al. 2015, 2017; Glover & Clark 2016; Clark et al. 2019a; Papadopoulos, Bisbas & Zhang 2018; Gong et al. 2020; Bisbas, Tan & Tanaka 2021).

In the distant Universe, additional problems arise. High-redshift galaxies are often observed solely in high-*J* CO lines (J = 3-2 and higher), due to the observational challenge of observing the two low-*J* CO lines.<sup>2</sup> Using the high-*J* lines means that global CO(J + 1, J)/(1–0) ratios must be assumed before an  $\alpha_{CO}$  factor can be used; given the

<sup>2</sup>Prior to the commissioning of its bands 1 and 2, low-*J* lines from high-redshift galaxies are inaccessible to the Atacama Large Millimetre Array

wide range of CO spectral-line energy distributions (SLEDs) found for LIRGs for J = 3-2 and higher (Papadopoulos et al. 2012b; Greve et al. 2014; Kamenetzky et al. 2016), these assumptions come with large uncertainties. Finally, at the highest redshifts ( $\gtrsim$ 4), low-*J* CO lines (and dust emission) can be severely suppressed for cold gas (and dust) reservoirs due to their low contrast against the ambient, rest-frame cosmic microwave background (da Cunha et al. 2013; Zhang et al. 2016).

In principle, radiative transfer models of well-sampled CO (and <sup>13</sup>CO) SLEDs can yield  $\alpha_{CO}$  values appropriate for a particular galaxy (or even galaxy class) (e.g. Papadopoulos et al. 2012b, 2014; Harrington et al. 2021). Nevertheless, the size of the CO line data sets per galaxy required to do this make it impractical (in terms of telescope time) to obtain  $M(H_2)$  for large galaxy samples. Amassing a large sample typically means only one or two lines can be gathered per galaxy, and thus a calibration of  $\alpha_{CO}$  and its uncertainties remains very valuable. The only practical way to achieve this is to cross-calibrate against the other galaxy-scale H<sub>2</sub> mass tracers.

Large-area far-infrared (FIR) and submillimetre (submm) surveys (e.g. Armus et al. 2009; Eales et al. 2010; Vieira et al. 2010; Kennicutt et al. 2011; Oliver et al. 2012; Hodge et al. 2013) ushered in a new era in which submm continuum emission from dust has been used widely as an alternative tracer of  $M_{\rm H2}$ , although it has been clear that submm-derived dust masses ( $\propto L_{850}$ ) and CO-derived molecular gas masses ( $\propto L'_{\rm CO}$ ) are tightly correlated ever since the first statistical submm survey of 100 local FIR-bright galaxies (SLUGS; Dunne et al. 2000). The first suggestions to use dust as an alternative to CO at high redshift (e.g. Santini et al. 2010; Magdis et al. 2012; Scoville et al. 2014) were followed quickly by work demonstrating its potential (e.g Scoville et al. 2016; Hughes et al. 2017; Orellana et al. 2017).

An advantage of using submm continuum emission from dust as an H<sub>2</sub> gas tracer is that it becomes easier to measure at high redshift, because of the negative *K*-correction (e.g. Blain & Longair 1993), while recent technological advances made it possible to image areas large enough to be free of cosmic variance, leading to the FIR/submm detection of many thousands of galaxies by the *Herschel Space Observatory*, for example. The use of dust as a gas mass proxy requires an estimate of metallicity, since the dust-to-gas ratio,  $\delta_{GDR}$ , is roughly proportional to metallicity (e.g. Muñoz-Mateos et al. 2009; Magdis et al. 2012; Sandstrom et al. 2013; Draine et al. 2014). The appropriate  $\delta_{GDR}$  can then be applied (e.g. Valentino et al. 2018). Whilst this requirement is often raised as a problem regarding the use of dust as a gas mass tracer, its dependence on metallicity is in fact weaker than that of CO.<sup>3</sup>

For galaxies selected at FIR/submm/mm wavelengths, it is safe to assume that the metallicity will be high, such that  $\delta_{GDR}$  will be broadly similar to those found for local metal-rich spirals and the Milky Way (Dunne & Eales 2001; Draine 2009; Magdis et al. 2012; Sandstrom et al. 2013; Rowlands et al. 2014; Yang et al. 2017; Berta et al. 2021). A detailed discussion of the advantages and

<sup>&</sup>lt;sup>1</sup>Because the CO(1–0) line is typically optically thick, with  $\tau_{10} \sim 5$ –10 (e.g. Bryant & Scoville 1996; Papadopoulos et al. 2012a: their equation 11), the *effective* critical density is lower still:  $n_{\rm cr}(\beta_{10}) = \beta_{10}n_{\rm crit} \sim 40$ –80 cm<sup>-3</sup>, where  $\beta_{10} = (1 - e^{-\tau_{10}})/\tau_{10}$  is the line escape probability.

<sup>(</sup>ALMA). The Jansky Very Large Array (JVLA), the Australia Telescope Compact Array (ATCA), and the Greenbank Telescope (GBT), have in some cases been able to access the faint low-J ( $J_u \leq 2$ ) CO lines, but it requires huge amounts of observing time in the best available weather.

<sup>&</sup>lt;sup>3</sup>Moreover, since  $H_2$  cannot be traced (in bulk) by any of its own lines, regardless of which other tracer (X) is used (dust emission, CO, or <sup>13</sup>CO, or C1 line emission), it will always be necessary to assume a [X/H<sub>2</sub>] abundance in order to proceed to a final  $H_2$  gas mass estimate.

disadvantages of using dust as a tracer of gas can be found in Genzel et al. (2015) and Scoville et al. (2017).<sup>4</sup>

A third method of tracing molecular gas – the use of atomic carbon lines – has come to the fore since ALMA became operational. Its promise was recognized by Papadopoulos, Thi & Viti (2004) and its first application as a tracer for molecular gas mass in galaxies gave good results (Weiß et al. 2003; Papadopoulos & Greve 2004), implying that: (a) the  $[CI]({}^{3}P_{1}-{}^{3}P_{0})$  lines are optically thin for the bulk of H<sub>2</sub> gas (Pérez-Beaupuits et al. 2015) and (b) atomic carbon is present throughout CO-rich molecular cloud volumes.

The latter contradicts the earlier simple plane-parallel PDR model where atomic carbon (and its line emission) occupied only a thin layer, sandwiched between C<sup>+</sup> in the outer and CO in the inner regions of FUV-illuminated molecular clouds (Tielens & Hollenbach 1985). However, observations have repeatedly shown excellent concomitance of CI line emission with CO line emission, by area and by velocity, and CI shows a tighter correlation with <sup>13</sup>CO than with <sup>12</sup>CO. CI is now thought to arise from same volume as the CO, with similar excitation conditions (e.g. Plume et al. 1999; Ikeda et al. 2002; Schneider et al. 2003; Beuther et al. 2014; Pérez-Beaupuits et al. 2015). Moreover, it may be that CI lines can also trace COdark molecular gas, should such phase exist in galaxies in significant amounts, e.g. due to CR-induced dissociation of CO to C (and O) (Bisbas et al. 2015).

Despite being much fainter than the C<sup>+</sup> line at 158 µm (the prime ISM cooling line), atomic carbon lines do hold certain advantages, namely: (a) they solely trace H<sub>2</sub> gas, whereas the C<sup>+</sup> line also traces the HI and HII gas reservoirs, which can be significant, especially in metal-poor systems (e.g. Madden et al. 1997; Liszt 2011; Papadopoulos & Geach 2012; Pérez-Beaupuits et al. 2015; Clark et al. 2019a); (b) the CI lines can remain excited for cold gas (e.g. for [C1](1–0):  $E_{10}/k_B \sim 24 \text{ K}$ ) unlike the C<sup>+</sup> line, where the  $\Delta E/k_B \sim 92 \text{ K}$  will keep it very faint for cold gas; (c) the frequencies of the two CI lines, at 492 and 809 GHz, remain accessible for galaxies over a much larger redshift range (and thus cosmic volume) than the C<sup>+</sup> line. In the latter case, its rest-frame frequency,  $\nu$ (C<sup>+</sup>)  $\sim$  1.9 THz, means the C<sup>+</sup> line is observable by ALMA's most sensitive receivers only at  $z \gtrsim 4$ .

Nevertheless, the high rest-frame frequencies of the C I line made early observations (and thus any calibration efforts) in the local Universe very difficult. Initially there had been relatively little observational work outside of the Milky Way, largely confined to extreme systems such as quasars and starburst nuclei (e.g. White et al. 1994; Weiß et al. 2005; Walter et al. 2011). These studies advocated a higher carbon abundance for these extreme systems,  $X_{\rm CI} = [{\rm C}^0/{\rm H_2}] = 5 12 \times 10^{-5}$ , compared to the  $X_{\rm CI} = 1-2.5 \times 10^{-5}$  seen in the Milky Way (Frerking et al. 1989).

More recently, *Herschel* observed many local galaxies in C I, although the [C I](1–0) line was at the edge of the observable range for the *Herschel* Fourier Transform Spectrometer (FTS), such that the sensitivity was somewhat compromised. As a result, most of the detected galaxies were either ULIRGs, starbursts, or low-metallicity dwarfs (Kamenetzky et al. 2014; Rosenberg et al. 2015; Lu et al. 2017; Jiao et al. 2017). A small sample of normal disc galaxies was mapped in C I (Jiao et al. 2019, hereafter J19; see also Crocker et al. 2019). J19 studied the spatial distribution of  $L'_{CI}$  and  $L'_{CO}$  at a ~1-kpc scale in 15 local galaxies. They concluded that C I is a good tracer of molecular gas, in the sense that it correlates well with CO and the

ratio  $L'_{\rm CI}/L'_{\rm CO}$  is distributed smoothly across galaxies. Comparing against CO(1–0) maps and the independent estimates of  $\alpha_{\rm CO}$  from Sandstrom et al. (2013), these resolved studies suggested  $X_{\rm CI} = 1.3-2.5 \times 10^{-5}$ , similar to the range in the Galaxy, and that found by the absorber study of Heintz & Watson (2020).

With ALMA now in routine operations, studies of CI have expanded to a broader range of galaxies, with a greater variety of average ISM conditions, over a wider range of redshift. These include SMGs, which lie mainly at z > 1 (e.g. Alaghband-Zadeh et al. 2013; Bothwell et al. 2017; Popping et al. 2017; Oteo et al. 2018; Nesvadba et al. 2019; Dannerbauer et al. 2019; Gómez-Guijarro et al. 2019), and MS galaxies at z = 0.35-1.2 (Valentino et al. 2018, 2020; Bourne et al. 2019; Dunne et al. 2021). CI has even been detected in the intracluster medium of the Spiderweb galaxy cluster at z = 2.16, as well as in several of its individual galaxies (Emonts et al. 2018). Routine use of CI as a tracer of molecular gas is currently limited by the lack of calibration studies to explore and determine the values and behaviour of the parameters involved, i.e.  $X_{\rm CI}$  and  $\alpha_{\rm CI}$ .  $X_{\rm CI} = 3 \times 10^{-5}$  has been adopted by almost all recent studies, taken from Weiß et al. (2003), determined from a comparison of analyses of CO and CI in the centre of M 82, which has unusually high  $[C^0/CO] \sim 0.5$ , whereas attempts to estimate  $X_{CI}$ in other ways - e.g. from absorption studies of Gamma-ray bursts and quasar absorbers (Heintz & Watson 2020) - have found lower values, consistent with the range seen in the Milky Way.

This paper presents the first dedicated cross-calibration study of the dust, <sup>12</sup>CO(1–0) and [CI](<sup>3</sup> $P_1$ –<sup>3</sup> $P_0$ ) emission in a sample of 407 galaxies from the literature, including MS galaxies and SMGs, such that we can compare their properties and tracer-(H<sub>2</sub> mass) conversion factors. We include the 250-µm-selected galaxies at z =0.35 observed with ALMA in all three tracers by Dunne et al. (2021) where our method was first briefly presented.

In Section 2, we describe the samples used in this analysis, the observables, and the derived quantities. In Section 3, we describe the Bayesian approach for producing optimized, self-consistent tracer-(H<sub>2</sub>-mass) conversion parameters between multiple tracers simultaneously. We then examine correlations of the observables to look for trends in Section 4. In Section 5, we investigate the trends we have found in the conversion factors and provide refined calibration recipes. Finally, in Section 6, we discuss the results and highlight the open questions. Throughout, we use a cosmology with  $\Omega_m = 0.27$ ,  $\Omega_{\Lambda} = 0.73$ , and  $H_0 = 71 \text{ km s}^{-1} \text{ Mpc}^{-1}$ .

## 2 DERIVING OBSERVATIONAL QUANTITIES

#### 2.1 Sample

The samples used in our study are those available in the literature – up-to-date as of early 2022 – which have at least two of the three tracers: submm dust continuum emission at  $\lambda_{rest} > 500\mu$ m,  ${}^{12}$ CO(1–0) or (2–1), and [C I]( ${}^{3}P_{1}-{}^{3}P_{0}$ ). Summarizing: 326 galaxies have both CO and submm continuum detections; 140 have both C I and submm dust continuum detections; 109 have both C I and CO detections; 101 have all three tracers. The sample covers the redshift range 0 < z < 6, and includes galaxies lying within 1 dex of the MS as well as extreme starbursts such as local ULIRGs and most high-z submm-selected galaxies. Full details and references are listed in Table 1. Lensed galaxies are included only where there is an estimate of the magnification,  $\mu$ , and all luminosities have been corrected by the magnification factor. Our sample includes the galaxies from one of the most comprehensive studies of dust as a tracer of molecular gas

<sup>&</sup>lt;sup>4</sup>Continuum dust emission does not yield information on kinematics, unlike spectral lines.

Sample name	Selection $\lambda_{obs}$ ( $\mu$ m)	Z	N <sub>CO</sub>	N <sub>C I</sub>	N <sub>sub</sub>	Notes	SF mode	References (see below)
high-z SMG	850-2000	2-6	89	42	114	Corrected for lensing	Both	а
Local SF		0	35	19	35	C I from FTS	MS	b
(U)LIRGs	60	0	85	19	114	C I from FTS	Both	С
z = 1	850	1	11	18	9	CO(2-1)	MS	d
z = 0.35	250	0.35	12	12	12		MS	е
0.04 < z < 0.3	160	0-0.3	48	0	54	VALES	MS	f

**Table 1.** Samples used for our comparisons.

*Notes*. In columns 4–6, *N* refers to the number of detections in each of the tracers.

*a*: Chapman et al. (2005, 2010), Weiß et al. (2005, 2013), Coppin et al. (2006), Hainline et al. (2006), Kovács et al. (2006), Daddi et al. (2009), Wu et al. (2009), Carilli et al. (2010, 2011), Engel et al. (2010), Harris et al. (2010), Ivison et al. (2010, 2011, 2013), Frayer et al. (2011, 2018), Riechers et al. (2011, 2013, 2020), Walter et al. (2011, 2012), Cox et al. (2011), Danielson et al. (2011), Lestrade et al. (2011), McKean et al. (2011), Magnelli et al. (2012), Thomson et al. (2012), Alaghband-Zadeh et al. (2013), Bothwell et al. (2013, 2017), Bussmann et al. (2013, 2015), Emonts et al. (2013), Sharon et al. (2013, 2016), Cooray et al. (2014), Messias et al. (2014, 2019), Negrello et al. (2014, 2017), Swinbank et al. (2014), Tan et al. (2014), Cañameras et al. (2015), Dye et al. (2015), Aravena et al. (2016), Scoville et al. (2017), Spilker et al. (2017), Oteo et al. (2017, 2018), Popping et al. (2017), Falgarone et al. (2017), Wong et al. (2017), Yang et al. (2017, 2019), Béthermin et al. (2018), Enia et al. (2018), Pavesi et al. (2019), Kaasinen et al. (2019), Leung et al. (2019), Nesvadba et al. (2018), Dannerbauer et al. (2019), Gómez-Guijarro et al. (2019), Jin et al. (2020), Drew et al. (2020), Neri et al. (2020), Harrington et al. (2021).

*b*: Mirabel et al. (1990), Tinney et al. (1990), Young et al. (1995), Casoli et al. (1996), Zhu et al. (1999), Curran, Aalto & Booth (2000), Dunne et al. (2000), Dunne & Eales (2001), Gao & Solomon (2004), Thomas et al. (2004), Stevens, Amure & Gear (2005), Albrecht, Krügel & Chini (2007), Kuno et al. (2007), Ao et al. (2008), Baan et al. (2008), Young, Bureau & Cappellari (2008), Galametz et al. (2011), Koda et al. (2011), Iono et al. (2012), Pappalardo et al. (2012), Schruba et al. (2012), Alatalo et al. (2013), Pereira-Santaella et al. (2013), Wong et al. (2013), Ueda et al. (2014), Liu et al. (2015), Rosenberg et al. (2015), Bolatto et al. (2017), Cao et al. (2017), J19, Jiao, Gao & Zhao (2021), Clark et al. (2018), Valentino et al. (2018, 2020), Hunt et al. (2019), Lapham & Young (2019), Sorai et al. (2019);

c:Dunne et al. (2000), Yao et al. (2003), Gao & Solomon (2004), Wilson et al. (2008), Chung et al. (2009), Papadopoulos (2010), Papadopoulos et al. (2012a), García-Burillo et al. (2012), Alatalo et al. (2016), Chu et al. (2017), Jiao et al. (2017), Lu et al. (2017), Yamashita et al. (2017), Herrero-Illana et al. (2019), Michiyama et al. (2020), Izumi et al. (2020);

d: Valentino et al. (2018, 2020), Bourne et al. (2019);

e: Dunne et al. (2021);

f: Villanueva et al. (2017), Hughes et al. (2017).

across cosmic time – Scoville et al. (2016), henceforth  $Sco16.^5$  The Scoville et al. sample has been updated as described in Appendix A.

In order to test for the effect of SF intensity or 'SF-mode' on any later results, we divide the sample into two groups, referred to hereafter as 'MS galaxies' and 'SMGs', the names of the groups are not meant to be accurate definitions but rather a reference to familiar categories. For this heterogeneous data-set, defining a simple criterion for two groups is not possible, and even if it were, a fuzzy boundary would still remain due to measurement errors and the inability to capture the complexity in a single parameter. The extreme starburst 'SMG' group contains the high-redshift submillimeter selected galaxies which were discovered in the pre-ALMA era and as such are extreme star forming systems (else they could not have been detected), plus the local ULIRGs and some LIRGs which have evidence for very intense and obscured regions (e.g. NGC 4418, IC 860) where conditions are likely to be extreme (Díaz-Santos et al. 2017; Falstad et al. 2021). The 'MS galaxy' group contains the lower luminosity local disc galaxies plus the LIRGS which are not extreme, the intermediate redshift sources selected at 250  $\mu$ m from the *Herschel*-ATLAS – z = 0.35 galaxies from Dunne et al. (2021) and the z < 0.3 VALES galaxies (Hughes et al. 2017), the  $z \sim 1$ galaxies (Valentino et al. 2018; Bourne et al. 2019) and the ASPECs sources denoted as 'MS' in that survey (Boogaard et al. 2020). (Full references are provided in Table 1.)

There are two situations where corrections to luminosities may be required, they are discussed below.

## <sup>5</sup>Although lensed galaxies were included in their work, the luminosities were not de-magnified.

#### 2.1.1 H1-dominated galaxies at low LIR.

For galaxies with a large fraction of H I within their optical disc, their dust tracing H I rather than H<sub>2</sub> makes a significant contribution to the submm continuum emission. Since our intention is to provide a calibration for H<sub>2</sub> rather than total gas, we apply a correction to  $L_{850}$  for galaxies with  $f_{\rm HI} = {\rm H~I/H_2} > 1$ , as described in Appendix B1. Galaxies corrected in this way are shown as cyan diamonds in the plots.

#### 2.1.2 Local galaxies mapped in C1 by the Herschel FTS.

The local galaxies mapped using the *Herschel* FTS by J19 present some complex issues. Some do not have CI and dust continuum measurements in matched apertures, and those same galaxies are often only detected in CI in the inner few kpc of the galaxy, where the ratios of  $L'_{CI}/L'_{CO}$  may also be biased – for example, by a lower  $\alpha_{CO}$  in galaxy centres. We discuss the issues in more detail in Section 4.4 and Appendix B2. Galaxies requiring a significant correction (>0.1 dex) to  $L'_{CI}$  are labelled as CI<sup>cor</sup>; they are shown in the plots as pink diamonds, but not included in the analysis unless specified.

#### 2.2 Observables

We will compare three tracers of molecular gas, where the observables (the luminosities  $L_{850}$ ,  $L'_{CO}$ , and  $L'_{CI}$ ) are empirically related to the molecular gas mass as

$$M_{\rm mol} = L_{850} / \alpha_{850} = \alpha_{\rm CO} L'_{\rm CO} = \alpha_{\rm CI} L'_{\rm CI}.$$
 (1)

## 966 L. Dunne et al.

The goal of this analysis is to determine self-consistent conversion factors  $\alpha_{850}$ ,  $\alpha_{CO}$  and  $\alpha_{CI}$  and study the physical properties they depend on, e.g. C abundance, gas-to-dust ratio ( $\delta_{GDR}$ ), dust emissivity. Our definition of the 'observables' is intended to be independent of as many assumptions as possible. For CO and C I, we use *L*' as defined by Solomon & Vanden Bout (2005)

$$L' = \frac{3.25 \times 10^7}{v_{\text{rest}}^2} \left(\frac{D_{\text{L}}^2}{1+z}\right) \left[\frac{\int_{\Delta V} S dv}{\text{Jy km s}^{-1}}\right] \quad \text{K km s}^{-1} \,\text{pc}^2, \tag{2}$$

where  $\int_{\Delta V} S dv$  is the velocity-integrated line flux density,  $D_L$  is the luminosity distance (Mpc), and  $v_{\text{rest}}$  is the rest frequency<sup>6</sup> of the transition in GHz.

Most of the galaxies in our compilation have been observed in the  ${}^{12}\text{CO}(J = 1-0)$  transition. However, some observations at high redshift target the  ${}^{12}\text{CO}(J = 2-1)$  line. We convert  $L'_{21}$  to  $L'_{10}$  using the line luminosity ratio  $R_{21} = 0.8$ ; if instead we were to set  $R_{21}$  to unity, this would not affect any of our conclusions. We do not use  $J \ge 3$  CO lines because the uncertainties in the global excitation corrections become too large for a useful calibration study.

We use only the C I  ${}^{3}P_{1}$ - ${}^{3}P_{0}$  line, as it is the least sensitive to the average excitation conditions, and correlates better with the low-*J* CO emission (Jiao et al. 2017, 2019; Crocker et al. 2019). Moreover, there is now evidence of strongly sub-thermal excitation for C I(2–1) (Harrington et al. 2021; Papadopoulos, Dunne & Maddox 2022), making it difficult to use this line as an H<sub>2</sub> mass tracer since its excitation is extremely uncertain.

For the dust continuum emission, we use  $L_{850}$ , the luminosity at rest-frame 850  $\mu$ m

$$L_{850} = 4\pi S_{\nu(\text{obs})} \times K\left(\frac{D_{\text{L}}^2}{1+z}\right) \quad \text{W}\,\text{Hz}^{-1},\tag{3}$$

where  $D_{\rm L}$  is the luminosity distance,  $S_{\nu({\rm obs})}$  is the observed flux density and *K* is the *K*-correction to rest-frame 850  $\mu$ m, defined as

$$K = \left(\frac{353 \,\text{GHz}}{\nu_{\text{rest}}}\right)^{3+\beta} \left(\frac{e^{h\nu_{\text{rest}}/kT_{\text{d}}} - 1}{e^{16.956/T_{\text{d}}} - 1}\right).$$
(4)

Here,  $v_{\text{rest}} = v_{\text{obs}}(1 + z)$ ,  $T_{\text{d}}$  is the luminosity-weighted dust temperature, from an isothermal fit to the spectral energy distribution (SED) with the dust emissivity,  $\beta$ , allowed to vary between 1.8 and 2.0.

Sco16 assumed a  $T_{\rm d} = 25 \,\mathrm{K}$  and  $\beta = 1.8$ , respectively, to extrapolate (or K correct) their observed submm luminosities to rest-frame 850  $\mu$ m. We make full use of the available data to refine this procedure as follows: (1) With sufficient data, we fit the SED ourselves with  $\beta = 1.8$  and estimate the rest-frame 850- $\mu$ m luminosity directly from the SED fit. (2) Failing that, we use the reported  $T_{\rm d}$  in the literature to make the extrapolation from the longest wavelength measurement available. (3) For SMGs with insufficient data points to have had their SED fitted, we adopt their observed average,  $T_d = 38 \text{ K}$  (da Cunha et al. 2015). The bulk of the high-z samples now have observations between 2 and 3 mm with ALMA, as such the extrapolation to rest-frame  $850\,\mu\text{m}$  is small, even at the highest redshifts. The shortest rest-frame wavelengths we deal with are  $\lambda_r \sim 250 \,\mu\text{m}$  for sources at  $z \sim 2-3$  observed at 850  $\mu$ m, which require K-corrections in the range 50–140. However, the important consideration is the potential uncertainty in that Kcorrection, not its absolute value. We tried alternatively using the Sco16 method of assuming  $T_d = 25 \text{ K}$  to extrapolate to rest-frame

<sup>6</sup>Where we use  $v_{rest}$ , Solomon & Vanden Bout (2005) use  $v_{obs}$ , hence the different exponent for (1 + z) cf. their equation (3).



**Figure 1.**  $T_{\rm mw}$  measurements from SED fitting as a function of redshift (top),  $L_{\rm IR}$  (middle), and SED colour  $L_{\rm IR}/L_{850}$  (bottom). The best-fitting line and  $2\sigma$  error are shown as a blue line and shaded region, the grey-dashed horizontal line shows the value of  $T_{\rm mw} = 25 \,\text{K}$  used by Sco16. There is a significant correlation with all three observables, such that the average  $T_{\rm mw}$  increases with redshift (r = 0.64),  $L_{\rm IR}(r = 0.74)$ , and SED colour (r = 0.80). Fit parameters are listed in Table 4.



*Cross-calibration of global* H<sub>2</sub> *gas mass tracers* 967

**Figure 2.** Tracer luminosity ratios as a function of lensing, redshift, and SFR.  $p_{\text{KS}}$  is the probability of no difference from a two-sided KS test. Left-hand column:  $L_{850}/L'_{\text{CO}}$  – there are no indications of any bias due to lensing or redshift. The third panel shows the effect of high  $f_{\text{HI}}$  in MS galaxies (we have not applied the H I correction factor to show the difference in the raw ratio). The green-shaded region consists of MS galaxies with  $\log L'_{\text{CO}} > 8.9$  and  $f_{\text{HI}} < 1$ . The black line is for all galaxies with  $\log L'_{\text{CO}} < 8.9$ , including the lo-VALES galaxies suspected of having  $f_{\text{HI}} > 1$ . Galaxies known to have  $f_{\text{HI}} > 1$  are shown as the dotted line to illustrate the similarity. The MS sub-group in the lower panel excludes the  $f_{\text{HI}} > 1$  and lo-VALES galaxies. Middle column:  $L_{850}/L'_{\text{CI}}$  – there are no indications of bias due to lensing or redshift. The third panel shows the C1<sup>cor</sup> galaxies from J19 compared to the rest of the MS sub-group; even with the aperture correction applied to the C1 flux (Appendix 4.4), they have a significantly different distribution of observed ratios. In the lower panel, we exclude C1<sup>cor</sup> galaxies and find no difference between galaxies with high and low SFRs. Right-hand column:  $L'_{\text{CI}}/L'_{\text{CO}}$  – the highest redshift SMGs (z > 3) have higher  $L'_{\text{CI}}/L'_{\text{CO}}$  at marginal significance (p = 0.012), but there are only 11 galaxies at z > 3. The third panel shows once more the difference between the C1<sup>cor</sup> galaxies in both high and low luminosity bins. Even though the C1 and CO are measured in the same apertures, the C1<sup>cor</sup> galaxies have much lower  $L'_{\text{CI}}/L'_{\text{CO}}$ . The bottom panel shows a low significance difference between high and low SFR galaxies (p = 0.056), becoming significant when including the C1<sup>cor</sup> galaxies. Means and KS results are given in Table 5.

 $L_{850}$  and found a maximum difference of a factor 1.6, with the average being 1.15 times. The true uncertainty due to the SED sampling and *K*-correction will be smaller than this, as we know from our work in Section 2.3.1 and Fig. 4 that the dust temperatures in SMG are much higher than 25 K. We, thus, do not consider the extrapolation to rest-frame 850  $\mu$ m to be a significant source of uncertainty or bias in this analysis.

#### 2.3 Physical dependencies of gas mass tracers

#### 2.3.1 Dust-H<sub>2</sub> calibration

Large dust grains ( $a \sim 0.1 \,\mu$ m) in thermal equilibrium with their incident radiation field emit as a modified blackbody (MBB), where the emission is related to the mass of hydrogen as:

$$M_{\rm H2} = \frac{L_{\nu}}{4\pi B(\nu, T_{\rm mw})} \ \kappa_{\rm H}(\nu).$$
(5)

The two physical quantities needed to calibrate dust continuum emission as a tracer of gas are therefore  $T_{\rm mw}$  and  $\kappa_{\rm H}$ . Expressing equation (5) in astronomical units for  $\lambda = 850 \,\mu$ m, we can write

$$\frac{M_{\rm H2}}{[\rm M_{\odot}]} = 6.14 \times 10^{-14} \frac{\kappa_{\rm H}}{[\rm kg\,m^{-2}]} \frac{L_{850}}{[\rm W\,Hz^{-1}]} \left(\frac{24.5}{T_{\rm mw}}\right)^{-1.4},\tag{6}$$

where we have simplified the exponential term in the Planck function as  $\sim (24.5/T_{\rm mw})^{1.4}$  for  $17 < T_{\rm mw} < 30$  K.

The mass-weighted dust temperature,  $T_{mw}$ , is often lower than the luminosity-weighted dust temperature,  $T_d$ , as derived from an isothermal MBB fit to the dust SED, because warm dust outshines cold dust per unit mass. There is an excellent discussion of this in the Appendix to Sco16 which we will not repeat here (see also Dunne & Eales 2001).

To determine  $T_{\rm mw}$ , we require a multicomponent MBB fit to a wellsampled dust SED (e.g. Dunne & Eales 2001), or an SED fit using a model that allows a range of radiation field strengths, leading to a range of dust temperatures (e.g. Draine et al. 2007). These methods



**Figure 3.** Relationships between the observed luminosities. The blue line and shaded regions in each panel show the best fit and the  $2\sigma$  uncertainty (see Table 4 for parameters). Top left-hand panel:  $L_{850}-L'_{C0}$ . Cyan diamonds indicate those local galaxies which have  $f_{HI} > 1$  and have corrected values of  $L_{850}$ . The best fit is shown, including the H I-corrected set, but excluding the lo-VALES galaxies (peach circles with  $\log L'_{CO} < 8.9$ ; see Appendix B1). Choosing only galaxies with  $f_{HI} < 1$  does not change the results; the fit is consistent with a linear slope with a high degree of confidence. Top right-hand panel:  $L_{850}-L'_{C1}$ . The resolved C 1<sup>cor</sup> galaxies from J19 that have an aperture correction (Appendix A) applied to  $L'_{C1}$  in order to compare to  $L_{850}$  are shown as pink diamonds. The dotted blue line shows the fit when these galaxies are included. The solid blue line is the fit that excludes these galaxies, while the dashed blue line is the fit to the log  $L_{IR} > 1$  galaxies only. Bottom left-hand panel:  $L'_{C1}-L'_{C0}$ . The pink diamonds are the C 1<sup>cor</sup> galaxies, where now the C I and CO are from matched regions, however, they are still offset to lower  $L'_{C1}$  for a given  $L'_{C0}$ . The fit including the pink diamonds is steeper than linear, while excluding them (solid line) returns a linear slope. Bottom right-hand panel:  $\sigma'_{850}-L'_{C1}$ . We compare the 850- $\mu$ m emissivity ( $\sigma'_{850} = M_{dust} \times \kappa_{850}$ ) to  $L'_{C1}$ . This allows us to show dust data in the resolved galaxies from Jiao et al. (2021) (pink stars) measured in the same region as [C1](1–0). The dotted blue line shows the fit including the pink stars, the solid line shows the fit excluding the C1<sup>cor</sup> galaxies from Jiao et al. (2021) (pink stars) measured in the same region as [C1](1–0). The dotted blue line shows the fit including the pink stars, the solid line shows the fit excluding the C1<sup>cor</sup> galaxies from Jiao et al. (2021) (pink stars) measured in the same region as [C1](1–0). The dotted

give broadly consistent results. As a rule of thumb, the range of  $T_{\rm mw}$  in local star-forming galaxies is 15–25 K (Dunne & Eales 2001;Dale & Helou 2002; Draine et al. 2007; da Cunha, Charlot & Elbaz 2008; Hunt et al. 2015; Bendo et al. 2015; Clark et al. 2015), increasing to 25–30 K in luminous starbursts at higher redshifts (Rowlands et al. 2014; da Cunha et al. 2015).

As the dust ( $\alpha_{850}$ ) factor is only weakly dependent on the assumed temperature at rest-frame 850  $\mu$ m, Sco16 and others assumed a constant  $T_{\rm mw} = 25$  K. If instead the true  $T_{\rm mw}$  were to be 15 [30] K, the dust and gas mass would be under[over]estimated by a factor  $\sim 2 \times [1.3 \times]$ , which is overshadowed by the other uncertainties. On the other hand, failure to account for any *systematic* trend of  $T_{\rm mw}$  with another physical parameter can introduce or mask correlations of the conversion factors with that physical parameter.

We therefore explore the validity of assuming constant  $T_{\rm mw}$  by collating measurements of  $T_{\rm mw}$  from the literature (Dunne & Eales 2001; Hunt et al. 2019), and additionally making our own fits where possible. In Fig. 1, we show that there are indeed strong correlations of  $T_{\rm mw}$  with the observables, namely z,  $L_{\rm IR}$  and the SED colour,  $L_{\rm IR}/L_{\rm 850}$ . There is a clear difference between samples with low and high SFRs, with  $\langle T_{\rm mw}({\rm MS})\rangle = 23.0 \pm 0.4$  K while  $\langle T_{\rm mw}({\rm SMG})\rangle = 30.1 \pm 0.7$  K. We fit empirical relations for the correlations in Fig. 1 (see Table 4). Where there is no direct estimate of  $T_{\rm mw}$  from an SED fit, which is the case for two-thirds of galaxies, we use these empirical relations<sup>7</sup> to derive  $T_{\rm mw}$  for use in our subsequent analysis.

<sup>7</sup>We restrict the predicted  $T_{\rm mw}$  such that  $T_{\rm mw} \leq T_{\rm d}$ .



**Figure 4.** Histograms of (top panel) luminosity-weighted dust temperature  $(T_d)$  from MBB fits (bottom panel) mass-weighted dust temperature  $(T_{mw})$  from fits allowing for multiple dust temperatures. Means and KS test results are given in Table 5.

Table 2. Samples used in cross-calibration analysis.

Sample	Tracers present	Ν	Median log $L_{\rm IR}$
daX	Dust, CO, and CI	101 (90)	11.65 (11.77)
Xa	CO and CI	109 (97)	11.66 (11.88)
Xd	Dust and CI	140 (128)	11.88 (12.06)
ad	CO and dust	326 (240)	11.54 (12.07)

*Notes. N* is the size of the sample upon which the analysis has been performed, excluding those with uncertain and potentially large corrections – see Section 4.4. Values in parenthesis are the number of galaxies in the samples with log  $L_{\rm IR} > 11$  and their median log  $L_{\rm IR}$ .

Appendix H2 compares our approach, where we use individual estimates of  $T_{\rm mw}$ , to the adoption of a constant  $T_{\rm mw} = 25$  K, where we will discuss those findings in Section 5.

In their work on strongly lensed SMGs at high redshift, which includes dust continuum emission as a constraint in a large-velocity gradient (LVG) model, Harrington et al. (2021) find that  $T_{\rm d} \sim T_{\rm mw}$  for most SMGs, with both measures of temperature higher than the  $T_{\rm mw} = 25 \,\text{K}$  commonly used in the literature. To ensure consistency with our other estimates of  $T_{\rm d}$ , we fitted the Harrington et al. (2021)

Table 3. Summary of the parameters required to reproduce this analysis.

Quantity	Set	Ст	СО	Dust
Physical		X <sub>CI</sub>	α <sub>CO</sub>	κ <sub>H</sub>
Empirical		$\alpha_{CI}$	$\alpha_{\rm CO}$	α <sub>850</sub>
SX, α, κ	$\log L_{\rm IR} > 11$	0.082	0.1646	0.1339
	BL	0.1125	0.1436	0.1294
BL = baseline (ex	cludes C Icor and lo-VAL	ES galaxies)		
Pair	Set	Mean l		
		BL	$\log L_{\rm IR} > 11$	
$\alpha_{\rm CO} X_{\rm CI}$	Xa	$-4.400 \pm 0.020$	$-4.383 \pm 0.021$	
	daX	$-4.393 \pm 0.021$	$-4.373 \pm 0.022$	
$\alpha_{\rm CO}/\kappa_{\rm H}$	ad	$-2.769 \pm 0.015$	$-2.798 \pm 0.018$	
	daX	$-2.867 \pm 0.025$	$-2.875 \pm 0.027$	
κ <sub>H</sub> X <sub>CI</sub>	Xd	$-1.529 \pm 0.021$	$-1.509 \pm 0.021$	
	Xeb	$-1526 \pm 0.024$	$-1.498 \pm 0.024$	

Note. Values here can be used to reproduce our method and should be applicable to other metal-rich samples.  $s_{X, \alpha, \kappa}$  are the intrinsic scatter on the log conversion factors,  $X, \alpha$ , and  $\kappa$ . 'Mean log pair' are the means of the log combinations of calibration factors listed in the 'Pair' column, quoted with the standard error on the mean. We list in the second column the sample used to derive these means, both the sample with the largest number of pairs and also for daX, which provides our reference set. We provide numbers both for the BL galaxies (excluding those discussed in Section 4.4) and also those with log  $L_{\rm IR} > 11$ . The differences are not significant.

photometry with three simple models: (1) an isothermal optically thin MBB; (2) an MBB with variable optical depth and – where there were enough data – (3) a two-component MBB. In agreement with Harrington et al., we find that a single dust temperature adequately describes the SED of these galaxies, in contrast to lower redshift (U)LIRGs and normal galaxies which are better fit with multiple dust components ( $T_d > T_{mw}$ ) and/or fits with higher FIR optical depths. The temperatures from the Harrington et al. turbulence model correlate best with our isothermal  $T_d$  measurements for these galaxies, and the temperatures returned when allowing variable optical depth,  $T_d(\tau)$ , are always significantly higher than those from the Harrington et al. model. We therefore do not use optically thick fits to yield  $T_{mw}$  for our high-redshift SMGs. We instead use twocomponent SED fits to the lensed *Planck* sources and the handful of SMGs with sufficient data for the empirical relations shown in Fig. 5.

The other key physical parameter in the dust–H<sub>2</sub> conversion is  $\kappa_{\rm H}$ , which is a combination of the dust mass absorption coefficient ( $\kappa_{850}$ ) and  $\delta_{\rm GDR}$ , such that<sup>8</sup>  $\kappa_{\rm H} = \delta_{\rm GDR}/\kappa_{850}$ . Briefly,  $\kappa_{850}$  is sensitive to the grain composition and structure (amorphous; crystalline; coagulated; mantled), while  $\delta_{\rm GDR}$  is roughly proportional to metallicity and, for galaxies with metallicity within a factor 2 of  $Z_{\odot}$ , as expected for those in our samples, can be taken to be roughly constant, at  $\delta_{\rm GDR} = 100-150$  (Sodroski et al. 1997; Dunne et al. 2000; Dunne & Eales 2001; Draine et al. 2007; Muñoz-Mateos et al. 2009; Leroy et al. 2011; Planck Collaboration 2011; Sandstrom et al. 2013; Jones et al. 2017; De Vis et al. 2021).

Fortunately, observational measures of  $\kappa_{\rm H}$  are available, both for the Milky Way and for external galaxies, with values of  $\kappa_{\rm H}$ ~2000 kg m<sup>-2</sup> in the Milky Way's diffuse interstellar medium (ISM) and  $\kappa_{\rm H} \sim 800$  kg m<sup>-2</sup> in dense clouds. Appendix C discusses in more detail how it is measured, and Table C1 provides a comprehensive set of observational and theoretical values for  $\kappa_{\rm H}$  from the literature.

It is impossible to disentangle the effect of changing dust properties ( $\kappa_{850}$ ) from changes in  $\delta_{GDR}$  in observational determinations of  $\kappa_{H}$ . While the decrease in  $\kappa_{H}$  towards denser sightlines in the Milky Way is thought to be due to the dust grains coagulating in denser

<sup>&</sup>lt;sup>8</sup>Literature studies generally present the dust-H<sub>2</sub> conversion in terms of  $\delta_{GDR}$  for a fixed emissivity,  $\kappa_{850}$ . Given the mounting evidence that  $\kappa_{850}$  varies within our own (Remy et al. 2017; Ysard et al. 2015, 2018; Köhler, Ysard & Jones 2015) and other galaxies (e.g. Clark et al. 2019b, but see also Priestley & Whitworth 2020), we prefer to work with  $\kappa_{\rm H}$  to avoid projecting all the variation in the H<sub>2</sub>-dust conversion factor on to  $\delta_{\rm GDR}$ .

T 1 1 4	D ( C 1 )	ODD Cr. 1. r	· 11 · MO		1 1 1 1 1	
Table 4.	Parameters of robust	ODR fits between	variables using MC	MC, co-variant error	s and including inti	insic scatter, $\ln \lambda$ .

Log x	Log y	Group	т	С	ln λ	rs	р	Ν
$\frac{L'_{\rm CO}}{L'_{\rm CO}}$ $\frac{L'_{\rm CO}}{L'_{\rm CO}}$	$L'_{CI} \\ L'_{CI} \\ L'_{CI}$	$BL BL + C I^{cor}$ $\log L_{IR} > 11$	1.023 (0.029) 1.078 (0.026) 0.950 (0.035)	- 0.93 (0.29) - 1.49 (0.25) - 0.18 (0.37)	- 2.09 (0.09) - 2.03 (0.09) - 2.07 (0.10)	0.94 0.96 0.92		109 121 97
$L'_{\rm CI} \\ L'_{\rm CI} \\ L'_{\rm CI}$	$L_{850} \\ L_{850} \\ L_{850}$	$BL BL + C I^{cor} log L_{IR} > 11$	0.976 (0.024) 0.937 (0.020) 1.024 (0.030)	14.33 (0.23) 14.71 (0.19) 13.86 (0.30)	-2.11 (0.09) -2.08 (0.08) -2.17 (0.09)	0.95 0.96 0.94		140 152 128
$L'_{\rm CI}$ $L'_{\rm CI}$	$\sigma'_{850} \ \sigma'_{850}$	BL BL + C 1 <sup>cor</sup>	1.024 (0.025) 0.997 (0.021)	- 2.28 (0.24) - 2.01 (0.20)	- 2.05 (0.08) - 2.07 (0.08)	0.95 0.96		140 152
$L'_{\rm CO}$ $L'_{\rm CO}$ $L'_{\rm CO}$ $L_{\rm IR}$	$L_{850} \\ L_{850} \\ L_{850} \\ L_{C1}'/L_{C0}'$	BL $f_{\rm HI} < 1$ $\log L_{\rm IR} > 11$ $BL + C I^{\rm cor}$	1.003 (0.015) 1.002 (0.017) 0.983 (0.026) 0.071 (0.02)	13.42 (0.15) 13.43 (0.16) 13.63 (0.26) - 1.57 (0.23)	$\begin{array}{r} - 2.01 \ (0.05) \\ - 1.99 \ (0.05) \\ - 1.91 \ (0.06) \\ - 1.70 \ (0.09) \end{array}$	0.96 0.96 0.93 0.26	0.005	326 310 226 121
L <sub>IR</sub> T <sub>d</sub> T <sub>d</sub>	$L'_{CI}/L'_{CO}$ $L'_{CI}/L'_{CO}$ $L'_{CI}/L'_{CO}$	BL BL + C I <sup>cor</sup> BL	0.034 (0.02) 1.23 (0.23) 0.83 (0.28)	-1.11 (0.25) -2.60 (0.35) -2.0 (0.4)	- 1.70 (0.09) - 2.20 (0.12) - 2.00 (0.15)	0.09 0.31 0.15	0.34 0.12	109 115 103
$L_{\rm IR}/L_{850}$ $L_{\rm IR}$	$T_{\rm mw}$ $T_{\rm mw}$		0.216 (0.010) 0.070 (0.004)	3.90 (0.12) 0.60 (0.05)	- 3.76 (0.15) - 3.10 (0.10)	0.82 0.80		152 152
L <sub>IR</sub> L <sub>IR</sub> L <sub>IR</sub> L <sub>IR</sub>	$\alpha_{850}$ $\alpha_{CO}$ $\alpha_{CI}$ $X_{CI}$	BL BL BL BL	$\begin{array}{c} 0.045 \ (0.007) \\ 0.59 \ (0.09) \\ - \ 0.052 \ (0.010) \\ 0.028 \ (0.011) \end{array}$	12.271 (0.082) - 0.91 (1.10) 1.896 (0.124) - 5.136 (0.133)	- 3.36 (0.30) - 1.00 (0.50) - 3.90 (0.20) - 3.70 (0.23)	0.46 0.46 -0.48 0.29	0.008	230 230 82 82

*Notes.* y = mx + c fit parameters are given with  $1\sigma$  errors in parentheses. Parameters are calculated accounting for the errors in both *x* and *y* using the robust ODR described in Appendix I. Errors are sampled using the EMCEE MCMC sampler. Intrinsic scatter ( $\lambda$ ) is fitted as a third parameter.  $r_s$  is the Spearman rank correlation coefficient, and *p* is the probability, shown when p > 0.005. *N* is the number of galaxies in that regression. 'Group' defines the galaxies on which the regression is performed: BL = baseline (excludes C1<sup>cor</sup> and lo-VALES galaxies), while galaxies with  $f_{HI} > 1$  are corrected as described in Appendix B1.



**Figure 5.**  $L'_{CI}/L'_{CO}$  luminosity ratio (equivalent to  $\alpha_{CO}/\alpha_{CI}$ ) as a function of luminosity-weighted (peak SED) dust temperature (left-hand panel) and log  $L_{IR}$  (right-hand panel). The yellow shaded regions in the right-hand panel represents the  $1\sigma$  range of values found by Crocker et al. (2019) for resolved galaxies mapped with the *Herschel* FTS. The vertical red and blue bars show the  $1\sigma$  range of values for QSOs/SMGs and the Milky Way, respectively (Walter et al. 2011; Frerking et al. 1989). The best fit to all galaxies and  $2\sigma$  confidence region are shown as the blue line and shaded area (parameters are given in Table 4). The  $L'_{CI}/L'_{CO}$  ratio has a significant correlation with both  $T_d$  and log  $L_{IR}$  when all galaxies are included, but when excluding the C 1<sup>cor</sup> galaxies, the correlation all but disappears (blue dotted line).

environments – a process expected to increase their emissivity (e.g. Köhler et al. 2015) – there may also be some decrease in  $\delta_{GDR}$  if the gas is accreted into dust mantles or ices (i.e. grain growth). Both effects are to be expected (e.g. Jones et al. 2017; Jones 2018) and both act to decrease  $\kappa_{\rm H}$ . Counter to that, the higher estimates of  $\kappa_{\rm H}$  in the diffuse atomic phase (lowest  $N_{\rm H}$  sightlines at high latitudes) in the Milky Way may be due, in part, to a lower dust emissivity for grains without ice mantles, where only the refractory cores remain,

subjected to harsher ultraviolet (UV) irradiation. Additionally, there is likely a metallicity gradient at high latitudes, leading to a higher  $\delta_{\rm GDR}$ , further increasing  $\kappa_{\rm H}$ . There is, thus, a qualitative expectation that denser regions with higher metallicity will have higher dust emissivity,  $\kappa_{850}$ , and lower  $\delta_{\rm GDR}$ , producing a lower  $\kappa_{\rm H}$ . More diffuse regions with lower metallicity will move in the opposite direction. In Section 5, we find that we can constrain the *range* of  $\kappa_{\rm H}$ , at least, and therefore the combination of  $\delta_{\rm GDR}/\kappa_{850}$ .

#### 2.3.2 CI-H<sub>2</sub> calibration

Here, we introduce the two physical parameters the  $X_{CI} = [C^0/H_2]$ average abundance ratio and the average excitation factor  $Q_{10} = N_1/N_{tot}$ , pertinent to the use of C I as a tracer of H<sub>2</sub>. The relationship between  $M_{H2}$  and the 'observable' – [C I](1–0) line emission – is (in astronomical units)

$$M_{\rm H2}(\rm M_{\odot}) = \frac{0.0127}{X_{\rm CI} \ Q_{10}} \left(\frac{D_{\rm L}^2}{1+z}\right) \left[\frac{\int_{\Delta V} S_{\rm [CI](1-0)} dv}{\rm Jy\,km\,s^{-1}}\right],\tag{7}$$

with  $D_{\rm L}$  in Mpc and  $\int_{\Delta V} S_{\rm [CI](1-0)} \Delta v$  in Jy km s<sup>-1</sup>. Expressed in units of line luminosity, this becomes

$$M_{\rm H2}(\rm M_{\odot}) = \frac{9.51 \times 10^{-5}}{X_{\rm CI} \, Q_{10}} \, L'_{\rm CI}. \tag{8}$$

The excitation term,  $Q_{10}$ , describes the relative fraction of carbon atoms in the J = 1 state. Under general non-LTE conditions, it is a function of both gas density, n, and  $T_k$  and is derived analytically in the Appendix to Papadopoulos et al. (2004). A recent study of the [C I](2–1)/(1–0) line ratio (Papadopoulos et al. 2022) finds that the C I lines are both sub-thermally excited in the ISM of galaxies, with the [C I](2–1) especially so (see also Harrington et al. 2021). Thus, the LTE expressions for  $Q_{10}$  should not be used, nor will the C I line ratio produce an estimate of  $T_k$  (both methods having been widely used in the literature to date). Details for  $Q_{10}$  are in the Appendix D, but in summary we find the following:

(i) The [C I](1–0) excitation term,  $Q_{10}$ , is a non-trivial function of density and temperature, but for the range  $T_k \ge 20$  K and log  $n \ge 2.5$  – which is where the bulk of H<sub>2</sub> in star forming galaxies is thought to reside –  $\langle Q_{10} \rangle = 0.48 \pm 0.08$  where the 99-per cent confidence range is quoted (see Papadopoulos et al. 2022 and Fig. D1 for details).

(ii) Due to a slight superthermal behaviour, higher density, higher  $T_k$  conditions can produce similar or even lower  $Q_{10}$  than lower density, lower  $T_k$  conditions. This breaks any intuitive link between  $Q_{10}$  and the ISM conditions, i.e. we do not necessarily expect a higher  $Q_{10}$  in SMGs compared to MS galaxies (see Fig. D1).

(iii) As the [C1](2–1) line is even more strongly sub-thermally excited, its  $Q_{21} = N_2/N_{\text{tot}}$  factor varies strongly.<sup>9</sup> This is the main reason why our current study is restricted to the [C1](1–0) line.

As the  $[C1]({}^{3}P_{1}-{}^{3}P_{0})$  line is optically thin for most conditions expected in spiral discs (Weiß et al. 2005; Pérez-Beaupuits et al. 2015; Harrington et al. 2021), the relationship between  $L'_{C1}$  and  $M_{H2}$ is proportional to  $X_{C1}$  – the abundance of carbon atoms relative to H<sub>2</sub>. This dependence on abundance is as expected for any method that employs tracers of H<sub>2</sub> gas mass other than the H<sub>2</sub> lines themselves.<sup>10</sup>

With the excitation factor  $Q_{10}$  varying no more than 16 per cent over the typical range of H<sub>2</sub> conditions in galaxies ( $T_k \ge 20$  K, log  $n \ge 2.5$ ), the major source of uncertainty in CI-based molecular gas mass estimates (and thus the major source of scatter in the  $\alpha_{CI}$  conversion factor) is the neutral carbon abundance,  $X_{CI}$ . The relatively recent introduction of the [CI](1–0) line as a gas tracer means that  $X_{CI}$  has not been widely explored – constraining it and investigating any potential trends is a key outcome of our cross-calibration work.

In the Milky Way,  $X_{CI}$  is found to vary only modestly, from  $0.8-2.2 \times 10^{-5}$  (e.g. Zmuidzinas et al. 1988; Frerking et al. 1989; Tauber et al. 1995; Ikeda et al. 2002), while a much higher value  $(X_{\rm CI} = 5 \times 10^{-5})$  has been inferred for the nearby starburst nucleus of M 82 (Schilke et al. 1993; White et al. 1994; Stutzki et al. 1997).<sup>11</sup> Thanks to ALMA, very high localized ratios of  $L'_{CI}/L'_{CO}$  (translating to high  $X_{\rm CI} = 5-7 \times 10^{-5}$ ) have also been measured in extreme regions, such as the Circum-Nuclear Disk (CND) of NGC 7469 which is believed to host an X-ray Dominated Region (XDR; Izumi et al. 2020) and the outflow region in NGC 6240 (Cicone et al. 2018). More modestly elevated  $L'_{CI}/L'_{CO}$  ratios tend to be found in the central nuclear regions of starburst galaxies (J19; Salak et al. 2019; Saito et al. 2020). However, when averaged over larger kpc scale regions the ratios become consistent with the average global ratios measured for this sample (see Fig. 5). Independent measurements of  $X_{CI} =$  $1.6^{+1.3}_{-0.7} \times 10^{-5}$  (for solar metallicity) were made by Heintz & Watson (2020) using UV absorption measures for a range of absorber systems across cosmic time. CRs (and X-rays) are expected to dissociate CO in favour of atomic carbon, increasing [C<sup>0</sup>/CO], a hypothesis supported by both simulations and observations (e.g. Bisbas et al. 2015; Clark et al. 2019a; Israel 2020; Izumi et al. 2020).

#### 2.3.3 CO-H<sub>2</sub> calibration

The <sup>12</sup>CO(1–0) line is optically thick in most (but not all, see e.g. Aalto et al. 1995) ISM conditions expected in galaxies. Unlike dust continuum emission where optical depths build up over large columns of dust, the entire CO line optical depth builds up within very small gas 'cells' (<0.1 pc) due to the very turbulent nature of the velocity fields, and the small thermal line widths (Tauber, Goldsmith & Dickman 1991; Falgarone et al. 1998). This localized nature of CO line optical depths and the macro-turbulent CO line formation mechanism allows a great simplification of the radiative transfer models of such lines, i.e. the use of the so-called Large Velocity Gradient (LVG) approximation. However, it also complicates the relationship between the CO line luminosity and the underlying H<sub>2</sub> gas mass, making the corresponding conversion factor,  $\alpha_{CO}$ , dependent on the thermal state of the gas, its average density, as well as its dynamic state.

Following Papadopoulos et al. (2012b), the  $\alpha_{CO}$  factor in an LVG setting is given by

$$\alpha_{\rm CO} = 2.65 \frac{\sqrt{n_{\rm H2}}}{T_{\rm b}} K_{\rm vir}^{-1} \left[ M_{\odot} \left( kkms^{-1} \, {\rm pc}^{2-1} \right] \right]$$
(9)

where  $n_{\rm H2}$  and  $T_{\rm b}$  are the average density (in cm<sup>-3</sup>) and the CO(1– 0) brightness temperature<sup>12</sup> for the molecular cloud ensemble while  $K_{\rm vir}$  describes the average dynamic state of the gas (self-gravitating clouds  $K_{\rm vir} \sim 1$ , unbound clouds  $K_{\rm vir} > 1$ ). In principle, multiphase LVG models of CO (and <sup>13</sup>CO) SLEDs can be used to constrain  $\alpha_{\rm CO}$ , but, in practice, this demands large line data sets per galaxy (e.g. Papadopoulos et al. 2014; Harrington et al. 2021), making it impractical for use in large galaxy samples. This is why in our study  $\alpha_{\rm CO}$  remains an empirical conversion factor to be (cross)-calibrated.

<sup>&</sup>lt;sup>9</sup>The  $Q_{21}(n, T_k)$  that enters the estimates of molecular gas mass when the [C1](2–1) line is used can vary almost by a factor of ~5, depending on  $(n, T_k)$ .

<sup>&</sup>lt;sup>10</sup>Even for optically thick tracers of H<sub>2</sub> gas, such as CO(1–0) line emission, a [CO/H<sub>2</sub>] abundance still enters the method via the CO–H<sub>2</sub> cloud volumefilling factor,  $f_{CO}$ , albeit not in a sensitive fashion unless a combination of strong FUV radiation and/or low metallicities selectively dissociate CO in the outer cloud layers while leaving the largely self-shielding H<sub>2</sub> intact (then  $f_{CO}$  can be  $\ll$ 1, see Pak et al. 1998 for details).

<sup>&</sup>lt;sup>11</sup>The measurement is in fact the [C<sup>0</sup>/CO] abundance, and a value for [CO/H<sub>2</sub>] has then to be assumed to infer  $X_{CI}$ .

<sup>&</sup>lt;sup>12</sup>Here, the cloud CO-H<sub>2</sub> volume filling factor is set  $f_{\rm CO} = 1$ .

## 972 L. Dunne et al.

## 2.3.4 Conversion factors and physical parameters

The two optically thin tracers – thermal dust continuum emission and CI – have a simple relation between the empirical 'mass-tolight' conversion parameter ( $\alpha_X$ ) and the physical conditions in the ISM (e.g. abundance, emissivity, temperature). We can write the empirical factors (equation 1) in terms of these physical parameters as follows<sup>13</sup>

$$\alpha_{850} = \frac{1.628 \times 10^{16}}{1.36 \,\kappa_{\rm H}} \left(\frac{24.5}{T_{\rm mw}}\right)^{-1.4} \,\rm W \, Hz^{-1} M_{\rm mol}^{-1} \,, \tag{10}$$

where the factor 1.36 corrects to total molecular mass, including He.

$$\alpha_{\rm CI} = 16.8 \left[ \frac{X_{\rm CI}}{1.6 \times 10^{-5}} \right]^{-1} \left[ \frac{Q_{10}}{0.48} \right]^{-1} \,\,\mathrm{M_{\odot}} \,\,\left(\mathrm{K\,km\,s^{-1}\,pc^{2}}\right)^{-1}.$$
(11)

Equation (11) also includes the factor 1.36 for He.

## **3 DERIVING SELF-CONSISTENT** CALIBRATION OF CONVERSION FACTORS

We next describe how we combine the measurements of multiple gas tracers in the most efficient way, in order to determine their crosscalibrations. Our goal is to find the empirical conversion factors ( $\alpha_{850}, \alpha_{CI}, \alpha_{CO}$ ) or physical parameters ( $\kappa_{H}, X_{CI}$ ), which produce a consistent estimate for  $M_{H2}$  in a given galaxy.

Our data set provides nested samples, each with a different set of available gas tracers. The daX sample has all three tracers available – dust continuum, CO, and C I, and contains 101 galaxies ( $N_{daX} = 101$ ). The names and statistics for the other samples are as follows: ad – CO and dust,  $N_{ad} = 326$ ; Xd – C I and dust,  $N_{Xd} = 140$ ; Xa – C I and CO,  $N_{Xa} = 109$ . The properties of these samples are in Table 2.

The best constraints at  $\log_{10} L_{IR} > 11^{14}$  come from the daX sample because it has three independent tracers of gas mass, but it lacks coverage of luminosities below  $l_{IR} = 11$ . The ad sample is the largest and spans the widest range in  $L_{IR}$ , reflecting the longer time for which CO observations have been possible for nearby galaxies.

We begin with the daX sample, to illustrate the method of optimization for the estimates of all three conversion factors simultaneously.<sup>15</sup> There are four unknowns namely:  $m = \log(M_{\rm H2})$ ,  $X = \log(X_{\rm C1})$ ,  $\kappa = \log(\kappa_{\rm H})$ , and  $\alpha = \log(\alpha_{\rm CO})$ , and three observables:  $L'_{\rm CO}$ ,  $L'_{\rm C1}$ , and  $L_{850}$ .

With an independent measure of the true  $M_{\text{H2}}$ , the observables would provide direct estimates of the three conversion factors; however, the value of  $M_{\text{H2}}$  is not known *a priori*, so we must use a probabilistic argument based on the fact the observations do provide constraints on the *relative* values of the conversion factors for each galaxy. There is, thus, a set of self-consistent conversion factors which link the observables to the true  $M_{\text{H2}}$ , with an unknown common constant factor.

The Bayesian approach we use is described, in detail, in Appendix F and requires an estimate of the intrinsic scatter for the logarithms of each of the factors:  $s_X$ ,  $s_\kappa$ , and  $s_\alpha$ . The observable luminosities relate to these factors as follows, where the coefficients

of proportionality are listed in Table 3:

$$\frac{L'_{\rm CO}}{L'_{\rm CI}} \propto \alpha_{\rm CO} X_{\rm CI}, \ \frac{L_{\rm 850}}{L'_{\rm CI}} \propto \kappa_{\rm H} X_{\rm CI}, \quad \frac{L'_{\rm CO}}{L_{\rm 850}} \propto \frac{\alpha_{\rm CO}}{\kappa_{\rm H}}.$$
(12)

We begin by measuring the intrinsic scatter between the three pairs of observables using an orthogonal distance regression (ODR) fitting method, which includes the intrinsic scatter,  $\lambda$ , as a third parameter in the analysis<sup>16</sup> (see Appendix I for full details). The three pair variances derived from the data are then used to estimate the intrinsic variance of the three individual conversion factors (the derivation can be found in Appendix E). The values of the intrinsic scatter for the parameters are given in Table 3, with  $X_{CI}$  having the smallest scatter between galaxies. This finding is purely empirical, requiring no assumptions about the values or trends of the conversion factors, and as such is very interesting.

As we do not have any independent measure of the gas mass with which to normalize our cross-calibration (four unknowns but only three measurements), we must make an assumption about the sample average of one of the physical or empirical conversion factors. However, with that assumption made transparent, the individual values can always be scaled to whichever normalization a reader wishes to adopt. *The relative values, however, are always the optimal solution*.

We choose to use the dust parameter  $\kappa_{\rm H} = \delta_{\rm GDR}/\kappa_{850}$  for this normalization, because there are no trends of  $L_{850}/L'_{\rm CO}$  or  $L_{850}/L'_{\rm CI}$  with  $L_{\rm IR}$  (see Figs 2 and G1) and  $\kappa_{\rm H}$  also has the best observational constraints.

For the Xa sample, where there are no dust continuum measurements, we normalize to  $X_{CI}^{N} = 1.6 \times 10^{-5}$ , which is the mid-range of the values suggested by the independent study of absorption lines by Heintz & Watson (2020).

All the galaxies in our sample are metal rich ( $0.5 < Z/Z_{\odot} < 2$ ) and so we assume that  $\kappa_{\rm H}$  ( $\delta_{\rm GDR}$ ) should be similar to that in the Milky Way and other local discs. Throughout the rest of this work, we will use as our reference point the mid-range of extragalactic determinations,  $\kappa_{\rm H} = 1500-2200 \,\rm kg \,m^{-2}$ , which are consistent with measurements of the diffuse ISM in the Milky Way. Our chosen normalization value, then, is  $\kappa_{\rm H}^{\rm N} = 1884 \,\rm kg \,m^{-2}$ , which is a good match to current theoretical dust models (THEMIS: Jones et al. 2017, and the updated Draine et al. 2007 modified by Hensley & Draine 2021).

For the standard Milky Way value of  $\delta_{GDR}$  (= 135, Magdis et al. 2012; Jones et al. 2017),  $\kappa_{\rm H} = 1884 \,\rm kg \,m^{-2}$  implies that  $\kappa_{850} = 0.071 \,\rm m^2 \,\rm kg^{-1}$ , similar to that used in many extragalactic studies (Dunne et al. 2000; James et al. 2002; da Cunha et al. 2013). For a  $\delta_{\rm GDR}$  fixed to 135, the range of  $\kappa_{\rm H}$  in extragalactic studies implies a range in  $\kappa_{850}$  of 0.06–0.09 m<sup>2</sup> kg<sup>-1</sup>. Table C1 lists  $\kappa_{\rm H}$  values from extragalactic and Galactic observations, as well as from theoretical dust models.

Note that the choice of normalization does not affect any of the trends, nor the ratio of the conversion parameters in the pairings; it merely sets the average value of the reference calibration parameter, to which the others are relative.

The sample mean expectation values for the other two parameters,  $\langle \alpha_{\rm CO} \rangle$  and  $\langle X_{\rm CI} \rangle$ , are next derived from our assumed value of  $\langle \kappa_{\rm H} \rangle$ , together with the mean ratios of the observables listed in Table 3. The *effective standard deviation* is also calculated – the intrinsic scatter

<sup>&</sup>lt;sup>13</sup>Hereafter, we omit the units for  $\alpha_{CO}$ ,  $\alpha_{CI}$ , and  $\alpha_{850}$ .

 $<sup>^{14}\</sup>mbox{Hereafter},$  we will refer to  $\log_{10}$  as simply log.

<sup>&</sup>lt;sup>15</sup>This method was first presented in brief in Dunne et al. (2021), where it was applied to the sample of z = 0.35 galaxies.

<sup>&</sup>lt;sup>16</sup>We need to multiply  $\lambda$  from the ODR fitting routine by  $\sqrt{2}$  because we need to know the intrinsic scatter of X - Y in our data set in order to determine the intrinsic scatter of each conversion factor in turn.

Cross-calibration of global H<sub>2</sub> gas mass tracers 973

of each parameter added in quadrature to the measurement error for that gas tracer. For example, for CO

$$\sigma_{\rm eff} = \sqrt{s_{\alpha}^2 + \sigma_{\rm CO}^2},$$

where  $s_{\alpha}$  is the intrinsic scatter in log( $\alpha_{CO}$ ), and  $\sigma_{CO}$  is the measurement error on log( $L'_{CO}$ ).

We can now estimate the probability of finding a particular set of conversion factors for any given galaxy. We use  $a_i$ , i = 1, 2, 3to denote the logarithms of the three conversion factors,<sup>17</sup> and write the mean expectation values and effective standard deviations as  $\langle a_i \rangle$  and  $\sigma_{i,\text{eff}}$ , respectively. Assuming that these follow Gaussian distributions, the probability of finding the factors,  $a_i$ , for any galaxy is

$$P \propto \prod_{i=1}^{N} \exp\left(-\frac{(a_i - \langle a_i \rangle)^2}{2\sigma_{i,\text{eff}}^2}\right)$$
$$= \exp\left(-\sum_{i=1}^{N} \frac{(a_i - \langle a_i \rangle)^2}{2\sigma_{i,\text{eff}}^2}\right).$$
(13)

Thus, the ratios of observable luminosities for any given galaxy can be used to determine the ratios of conversion factors (equation 12), and the common scaling factor that maximizes the probability in equation (13) is the best estimate of  $M_{\rm H2}$ . The derivation in Appendix F shows that this reduces analytically to a simple inverse variance weighted mean, such that

$$\log M_{\rm H_2}^{\rm opt} = \frac{\sum_{i=1}^{N} (m_i \times w_i)}{\sum_{i=1}^{N} w_i},$$
(14)

where  $w_i = 1/\sigma_{i,\text{eff}}^2$ , and  $m_i$  is the log mass estimate for each tracer.  $m_i = l_i + \langle a_i \rangle$ ,

where  $l_i$  is the measured observable (log luminosity) and  $\langle a_i \rangle$  is the sample mean expectation value for the conversion factor. Once the optimal mass is determined this way, we can then estimate the corresponding optimal conversion factor on a per-galaxy basis, as

$$a_i = m^{\text{opt}} - l_i. \tag{15}$$

The error on the optimal mass is simply the error on the inverse variance weighted mean

$$\sigma_m^{\text{opt}} = \left(\sum_{i=1}^N w_i\right)^{-1/2},\tag{16}$$

and the error on each of the conversion factors, accounting for co-variance is

$$\sigma_{ai} = \sqrt{\sigma_{\mathrm{m}^{\mathrm{opt}}}^2 + \sigma_{li}^2 \left(1 - \frac{2w_i}{\sum_{j=1}^N w_j}\right)},\tag{17}$$

where  $\sigma_{li}$  is the logarithmic measurement error on the observable quantity, e.g.  $L'_{CO}$ ,  $L'_{CI}$ ,  $L_{850}$ .

By design, each tracer for a given galaxy, together with its optimized conversion factors, will produce the same gas mass, such that  $M_{\rm H2}{}^{\rm CO} = M_{\rm H2}{}^{\rm CI} = M_{\rm H2}{}^{\rm dust}$ .

## **4 TRENDS IN THE LUMINOSITY RATIOS**

As our cross-calibration process relies on measurements of the luminosity ratios, it is first instructive to look at the trends in these

<sup>17</sup>For ease of representation,  $a_{850} = -\log(\alpha_{850})$ .

observables to better understand any subsequent trends in the derived conversion factors.

Histograms of the tracer ratios are shown in Fig. 2, split by factors such as lensing, redshift, SFR, and other notable quantities. The correlations of the three tracer luminosities are shown in Fig. 3, where the various samples are colour coded and labelled and in each panel the blue line and shaded region represent the best fit and  $2\sigma$ error interval. Fitting was performed using our own ODR method, which includes *x* and *y* errors, intrinsic scatter as a third parameter, and covariance in errors where required. The method is described in detail in Appendix I. Fit parameters, slope *m*, intercept *c*, and scatter ln  $\lambda$ , are listed in Table 4 and statistics for the various subsets from Fig. 2 are given in Table 5. It is instructive to look at these two plots together for the same luminosity pairs.

## 4.1 L<sub>850</sub> versus L'<sub>CO</sub>

The left-hand column of Figs 2 and 3(a) show the quantities  $L_{850}$ versus  $L'_{CO}$ . There are no significant differences in the distribution of  $L_{850}/L'_{CO}$  with lensing, redshift or SF-mode, but the observed uncorrected ratio is significantly higher for galaxies with log  $L'_{CO}$  < 8.9 (Fig. 2; left green histogram). These log  $L'_{CO} < 8.9$  galaxies tend to have optical discs dominated by atomic hydrogen ( $f_{\rm HI} > 1$ ) and the likely increased contribution to  $L_{850}$  from dust associated with H I rather than H<sub>2</sub> results in the offset to higher  $L_{850}/L'_{CO}$  ratios. For local galaxies with  $f_{\rm HI} > 1$ , we apply a correction (Appendix B1) which appears to remove this offset (cyan diamonds in Fig. 3a). Galaxies with log  $L'_{CO}$  < 8.9 from the VALES sample at 0.04 < z < 0.3 (Hughes et al. 2017; Villanueva et al. 2017) have noticeably higher  $L_{850}/L'_{CO}$  ratios than VALES galaxies with log  $L'_{CO} > 8.9$ (peach circles in Fig. 3a and Table 5). There are no published HI measurements for VALES, but we suspect that the lo-VALES sample with log  $L'_{CO}$  < 8.9 is likely to be H I-rich, based on the similarity between the log  $L'_{\rm CO}$  < 8.9 and  $f_{\rm H\,I}$  > 1 categories in the third panel. We therefore exclude lo-VALES from our averages, as we suspect they are in need of a correction for HI but we have no means to apply one. We also recommend that a low  $L'_{CO}$  requires careful consideration of HI-related dust. In Fig. 3(a), the tracers show a linear dependence, regardless of exactly which galaxies are included (Table 4). The lo-VALES galaxies are excluded from all the fits.

## 4.2 $L_{850}$ versus $L'_{CI}$

The central column of Figs 2 and 3(b) show  $L_{850}$  versus  $L'_{CI}$ , the pair with the least scatter ( $\ln \lambda$  in Table 4). There are no significant differences in the distributions of  $L_{850}/L'_{CI}$  as a function of lensing, redshift, or SF-mode. The  $L_{850}-L'_{CI}$  sample has 12 galaxies with large angular sizes from J19 that have only part of their disc detected in CI, denoted C I<sup>cor</sup>. To compare  $L'_{CI}$  and  $L_{850}$  for these galaxies, we need to apply an aperture correction to their CI fluxes, thereby assuming that their  $L'_{CI}/L'_{CO}$  ratios remain roughly constant across the disc (see Appendix 4.4). Some of the correction factors are very large (up to 0.7 dex); even after correction, their  $L_{850}/L'_{CI}$  ratios are significantly offset from the rest of the MS sample (green histogram). Fig. 3(b) shows  $L_{850}$  versus  $L'_{CI}$ , with the C I<sup>cor</sup> galaxies as pink diamonds. The fit to all galaxies including the C Icor subset is shown as a dotted blue line, and has a sub-linear slope  $m(3\sigma)$ , although the difference is very small ( $m = 0.937 \pm 0.020$ ). Excluding the C I<sup>cor</sup> galaxies from the fit leaves a linear relationship, shown by the blue solid line. At first glance, this and the green histogram in Fig. 2 suggests that our CObased corrections are insufficient; however, Fig. 3(d) shows that the problem is not simply the assumption used to correct  $L'_{CI}$  to match the

Table 5. Two-sample KS-test result and Z-test statistic for the following parameter pairs shown in Figs 2 and 4.

Quantity	Α	N <sub>A</sub>	В	$N_{\rm B}$	Ā	Ē	$Z(\sigma)$	P <sub>KS</sub>
$L_{850}/L'_{CO}{}^{a}$	$\log L'_{\rm CO} < 8.9$	50	$L'_{\rm CO} > 8.9 ({\rm MS})$	138	$13.600 \pm 0.030$	$13.420 \pm 0.016$	5.3	3e-5
$L_{850}/L'_{CO}{}^{a}$	$f_{\rm HI}$ < 1 (MS)	168	$f_{\rm HI} > 1 ({\rm MS})$	24	$13.456 \pm 0.014$	$13.685 \pm 0.042$	5.2	3e-6
$L_{850}/L'_{CO}{}^{a}$	lo-VALES	7	$f_{\rm HI} > 1$	17	$13.741 \pm 0.061$	$13.663 \pm 0.053$		0.57
$L_{850}/L'_{CO}$	$f_{\rm HI} < 1 \; ({\rm MS})$	168	$f_{\rm HI} > 1 \; ({\rm MS}^b)$	17	$13.456 \pm 0.014$	$13.448 \pm 0.039$		1.0
$L_{850}/L'_{CI}$	MS	61	C I <sup>cor</sup>	12	$14.108 \pm 0.027$	$14.36 \pm 0.036$	5.6	6e-4
$L'_{CI}/L'_{CO}$	z < 3 (SMGs)	37	z > 3	11	$-0.686 \pm 0.031$	$-0.512 \pm 0.044$	3.2	0.012
$L'_{CO}/L'_{CI}$	MS	55	SMGs	54	$-0.743 \pm 0.028$	$-0.651 \pm 0.028$	2.3	0.056
$L'_{\rm CO}/L'_{\rm CI}$	$MS + C I^{cor}$	66	SMGs	54	$-0.786\pm0.026$	$-0.651\pm0.028$	3.5	0.006
T <sub>d</sub>	MS	174	SMGs	160	$31.1 \pm 0.4$	$38.3 \pm 0.7$	8.8	2e-12
$T_{\rm mw}$	MS	82	SMGs	52	$23.0\pm0.4$	$30.1 \pm 0.7$	8.8	7e-13

*Notes.* <sup>*a*</sup>Using  $L_{850}$  without correction for  $f_{HI} > 1$  (as this is the driver of the difference).

<sup>b</sup>Not including the lo-VALES galaxies and with the H I correction applied.

global  $L_{850}$ . This plot shows the dust emissivity cross-section,  $\sigma'_{850}$ , equivalent to  $L_{850}$  but with the temperature sensitivity removed.<sup>1</sup> Importantly, this quantity is measured in the same aperture as  $L'_{CI}$ . The CIcor galaxies are shown as pink stars, and they - along with many other low-luminosity galaxies in J21 - still appear to have less CI emission for a given amount of dust.

## 4.3 $L'_{CI}$ versus $L'_{CO}$

The right-hand column of Figs 2 and 3(c) show  $L'_{CI}$  versus  $L'_{CO}$ . There are no significant differences in  $L'_{CI}/L'_{CO}$  for strongly lensed versus unlensed sources, but the highest redshift, z > 3, galaxies have higher  $L'_{CI}/L'_{CO}$  ratios at marginal significance (p = 0.012). There are only 11 galaxies at z > 3 and a larger sample is needed to determine if this is a genuinely significant trend. Fig. 3(c) shows the CI<sup>cor</sup> galaxies as pink diamonds, where the CI and CO fluxes are measured in the same apertures by J19. The solid line shows the fit to all galaxies, which is non-linear at  $3\sigma$  significance (m = $1.078 \pm 0.026$ ). The slope becomes linear once the C I<sup>cor</sup> galaxies are removed. The green histogram in Fig. 2 (right) shows more clearly why we see this: the C I<sup>cor</sup> galaxies have significantly lower  $L'_{CI}/L'_{CO}$ ratios compared to other galaxies at similar or higher luminosity. The bottom histogram shows a marginal difference between galaxies with different SFRs (p = 0.056) when excluding the C I<sup>cor</sup> galaxies, which becomes significant when they are included (p = 0.006).

#### 4.4 Resolved CI fluxes from Herschel FTS mapping

The local resolved galaxies observed with Herschel FTS (J19) lie off the global trends seen in Fig. 3. There are possible physical explanations why lower luminosity, and more quiescently starforming galaxies might have lower C I/CO line ratios (for example, different ISM environments in terms of their position in the CR energy density versus average molecular gas density diagram: see fig. 1 in Bisbas et al. 2015). Low ratios of  $L'_{CI}/L'_{CO}$  have been found in other studies, most intriguingly in the case of the interacting LIRG NGC 6052 using ALMA (Michiyama et al. 2020), and some high-z strongly lensed sources (Harrington et al. 2021). Such ratios tend to be unusual in higher luminosity samples, however, whereas the resolved FTS sample has a very low average for the CI line ratios with both CO and dust.

the same data but drew different conclusions (Crocker et al. 2019). We can therefore only note that the CI fluxes from Herschel FTS mapping data sets are not always consistent when analysed by different teams.<sup>19</sup> As these resolved FTS measurements are essentially the only source of CI data at log  $L_{IR} < 11$ , and carry a lot of weight in  $L_{\rm IR}$  and SFR correlations, we chose not to include the C I<sup>cor</sup> galaxies in the statistical analysis. If, instead, we take the J19 measurements at face value - they signpost a fundamental physical change in CI properties, a finding which clearly warrants further study with ground-based facilities. We discuss possible physical mechanisms for changes in the  $L'_{CI}/L'_{CO}$  and  $L'_{CI}/L_{850}$  ratios in Appendix B2. 4.5 Trends with global indicators of star formation

We recommend caution in the interpretation of the data for these resolved FTS sources because another team subsequently presented

Finally, we check to see if any of the tracer ratios are sensitive to SFR indicators. In our data set, the dust observables  $L_{IR}$  and  $T_d$  are indicators of the intensity and magnitude of star formation in galaxies (e.g. Kennicutt 1998; Foyle et al. 2012; Liu et al. 2021). As expected, the distribution of  $T_d$  is very different for the MS galaxies and SMGs (Fig. 4 and Table 5), reflecting the increase in the intensity of star formation in the SMGs. The only tracer ratio sensitive to these SF indicators is  $L'_{CI}/L'_{CO}$ , which in Fig. 5 is seen to increase with  $L_{IR}$  and  $T_{\rm d}$  when all galaxies are considered. Such a trend was not reported for smaller samples over a more limited range of luminosity (e.g. Jiao et al. 2017, 2019), presumably because of limited statistics. However, if the C1cor galaxies (pink diamonds) are excluded, the correlation all but disappears (blue dotted line).

Naively, we might expect  $L'_{CI}/L'_{CO}$  to rise with increasing SFR intensity, due to the expected destruction of CO by CR in high-SFR environments (e.g. Bisbas et al. 2015). For a given range of  $H_2$ densities in the typically hierarchical molecular clouds, any increased CR-induced ionization rate,  $\zeta_{CR}$  (due to a rising average CR energy density,  $U_{CR}$ ) will destroy CO in the lower density, more extended areas, while leaving CO still tracing H2 in the more compact, denser regions (see also fig. 1 of Papadopoulos et al. 2018 for a visual effect of this). Intriguingly, the gas density,  $n(H_2)$ , and CR ionization rate,

 $<sup>^{18}\</sup>sigma'_{850}$  is derived from the data provided in Jiao et al. (2021) by multiplying the dust mass in the CI aperture by the  $\kappa_{850}$  used in their method,  $\kappa_{850}$  $= 0.034 \text{ m}^2 \text{ kg}^{-1}$  (Draine 2003; Draine et al. 2007).

<sup>&</sup>lt;sup>19</sup>Crocker et al. did not provide integrated fluxes, nor a method to determine them from their published measurements; hence, we cannot use their work directly in our analysis. Q. Jiao has provided us with the maps used in J19, enabling us to check the measurements independently and extend our analysis, but we have had no responses to our requests for integrated fluxes or the details of the method used from the Crocker team.

 $\zeta_{CR}$ , will compete against each other in ULIRG/SMG environments, with the higher  $\langle n \rangle$  expected in their highly turbulent ISM tending to keep the ordinary CO/C I chemistry in place, even when exposed to the higher  $\zeta_{CR}$  values.<sup>20</sup> Guessing which one will win this highly non-linear competition (see fig. 1 and 8 in Bisbas et al. 2015) is dangerous in the absence of CO and C I line data. These effects have been probed with a variety of simulations (e.g. Bisbas et al. 2015, 2021; Clark et al. 2019a; Gong et al. 2020) and while showing similar trends, they are not easily parameterisable in terms of  $n(H_2)$  and  $\zeta_{CR}$ ; one reason why such cross-calibration efforts of the available gas mass tracers are so important.<sup>21</sup>

The other two tracer ratios show no trends with either  $L_{IR}$ ,  $T_d$  (our proxies for SFR) – we present the relevant plots in Appendix G1 for completeness.

## **5 RESULTS**

In this section, we present the results of the optimization method, firstly for the daX sample, for which we have all three gas tracers, and later for the other three samples, for which we have pairs of tracers. We investigate trends of the conversion factors with  $L_{IR}$  and SFR. Mean values for the conversion factors are listed in Table 6.

Fig. 6 shows the results for the daX sample (Figs 7–9 present the same results for each of the samples in turn). The top row of each plot shows the distribution of the relevant physical parameter for CI and dust, and the conversion factor for CO:  $X_{CI}$ ,  $\kappa_{H}$ , and  $\alpha_{CO}$ . The lower left-hand panels show the same quantities for the individual galaxies as a function of  $L_{\rm IR}$ ; each panel indicates a reference measure to give context. For  $X_{CI}$ , the horizontal lines indicate the measured extremes found in the local Universe: Orion A/B clouds in the Milky Way (Ikeda et al. 2002) and the starburst centre of M82 (White et al. 1994), while the grey shaded region shows the range of values inferred from observations of GRB hosts and QSO absorbers for solar metallicity by Heintz & Watson (2020). They use a method which does not rely on emission measures of dust, CO, or CI and so can be considered independent. For  $\alpha_{CO}$ , the horizontal lines indicate the typical  $\alpha_{CO}$  for the Milky Way (Bolatto et al. 2013) and that commonly adopted<sup>22</sup> for ULIRGs and SMGs (Downes & Solomon 1998). For  $\kappa_{\rm H}$ , we show a shaded band indicating the range derived for local galaxies (see Table C1), along with lines showing the value for the most diffuse and dense sightlines in the Milky Way (Remy et al. 2017). The right-hand lower panels show the running logmeans as a function of  $L_{IR}$  to make it easier to see any trends, and additionally includes<sup>23</sup> the empirical parameters,  $\alpha_{CI}$  and  $\alpha_{850}$ . The solid shaded bins are the means for the grey points, which are those used to determine the calibration; the yellow points are C I<sup>cor</sup> and the semitransparent pentagon is the mean of those - see Section 4.4 for more details.

Fig. 6 shows that for galaxies with log  $L_{IR} > 11$  there are only weak trends of the conversion factors with  $L_{IR}$ . While the normalization ( $\kappa_{H}^{N} = 1884 \text{ kg m}^{-2}$ ) was chosen to produce average dust properties

 $^{22}$  In this panel,  $\alpha_{CO}$  does not include the factor 1.36 for He.

 $^{23}As$  elsewhere, the empirical parameters  $\alpha_{C\,I}$  and  $\alpha_{850}$  include the factor 1.36 for He.

consistent with the Milky Way and other nearby spirals, the CO and C I conversion factors derived from the luminosity ratios also lie within the ranges expected from independent studies. The averages at log  $L_{\rm IR} < 11$  are based on only a small number of points (12) and more C I studies are required to probe quiescent local galaxies.

#### 5.1 A calibration for the gas masses

We next give a prescription for estimating gas mass, tailored to how many tracers are available and – where appropriate – the type of galaxy being investigated.

#### 5.1.1 Dual-band

While the information content is greatest for the daX sample, which has three tracer pairs to optimize, the method presented in Section 3 still improves the cross-calibration for samples which have two tracer measurements, i.e. one pair. The results for the Xd, Xa, and ad samples are shown in Figs 7–9 and behave similarly to the daX sample, as one would hope given that the daX galaxies are a subset of the others. The pink diamonds in the lower left-hand panels in Figs 7 and 8 denote the C1<sup>cor</sup> galaxies. The cyan diamonds in the lower left-hand panel of Fig. 9 are galaxies with  $f_{\rm HI} > 1$  which have been corrected for the contribution of dust mixed with the H I gas, as described in Appendix A. The open peach circles are the lo-VALES galaxies, which we suspect to have  $f_{\rm HI} > 1$  (see Section 4.1) but which we cannot correct. We do not include these in any averages or histograms.

The method previously used in the literature (e.g. Alaghband-Zadeh et al. 2013; Scoville et al. 2016; Hughes et al. 2017; Orellana et al. 2017; Valentino et al. 2018) has been to assume one tracer in a pair (e.g.  $L'_{CO}$ ) has a known conversion ( $\alpha_{CO}$ ), then to fix that factor for all galaxies in order to estimate the second (i.e. the one of interest). We show this simple method alongside our optimal method as grey lines and dashed grey error bars in the relevant panels of Figs 7-9. The scatter in the conversion factors for the optimized estimates are governed by the intrinsic scatter we inferred in our analysis of the data in Appendix E, free of assumptions. In contrast, the simple method proscribes that there is no scatter in the known conversion factor, and therefore all of the intrinsic scatter in the luminosity ratio is attributed to the second conversion factor of interest. The optimized method presented here does not assume an ad hoc preference for any particular conversion factor: as it is based on empirical variance analysis, it uses more of the available information to improve the accuracy of the estimated conversion factors. The histograms in Figs (7–9) show that the scatter in the factor of interest is larger when using the simple method, and the trends in the running medians are also more exaggerated.

Comparing the parameter estimates using three tracers to those using two tracers for the same galaxies allows us to test the accuracy of these two-tracer estimates. The details are in Appendix H, but in summary there is a reasonable correlation between the three-tracer and two-tracer estimates, without bias (Fig. H1) and an average scatter of 0.06–0.08 dex.

Thus, when multiple  $H_2$  tracers are available, we recommend the procedure outlined in the example below.

Example: Take the example of a galaxy with observations of both dust and CO. We take the mean value for the appropriate pair combination from Table 3:  $\alpha_{CO}/\kappa_{H} = 0.00133$ . For our adopted sample mean normalization of  $\kappa^{N} = 1884 \text{ kg m}^{-2}$ , we now infer the sample mean expectation value of  $\langle \alpha \rangle = 0.00133 \times 1884 = 2.5$ 

<sup>&</sup>lt;sup>20</sup>We here assume that CR energy density  $U_{CR} \propto \rho_{SFR}$  and CR ionization rate  $\zeta_{CR} \propto U_{CR}$ .

<sup>&</sup>lt;sup>21</sup>On an individual galaxy basis, one could assemble well-sampled CO, <sup>13</sup>CO, and C1 line SLEDs and overcome these problems with detailed analysis (e.g. Papadopoulos et al. 2014). However, even in the ALMA era, this remains very expensive in terms of telescope time making it prohibitive for large samples of galaxies.

Table 6.	Mean optimized	conversion	factors	for	our various	samples.
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Sample	Selection	Ν	$X_{CI}$ (× 10 <sup>-</sup>	$\sigma_{\bar{X}}$	$\alpha_{\rm CO}$ M <sub><math>\odot</math></sub> ( $\kappa$ km s <sup>-</sup>	$\sigma_{\bar{\alpha}}$	$\frac{\kappa_{\rm H}}{({\rm m}^2{\rm kg}^-)}$	$\sigma_{\bar{\kappa}}$	$\delta_{\rm GDR}$
daX	$\log L_{\rm IR} > 11$	90	$1.59^{+0.45}_{-0.28}$	0.04	$2.66^{+0.96}_{-0.70}$	0.10	1990+738	86	141
daX	$\log L_{\rm IR} < 11$	12	$1.18^{+0.60}_{-0.29}$	0.13	$2.44^{+0.56}_{-0.51}$	0.17	$1571^{+732}_{-525}$	163	112
Xd	$\log L_{\rm IR} > 11$	128	$1.59^{+0.47}_{-0.35}$	0.04	0.01		$1946_{-464}^{+654}$	53	138
Xd	$\log L_{\rm IR} < 11$	12	$1.24^{+0.57}_{-0.27}$	0.13			$1503^{+604}_{-369}$	145	107
ad	$\log L_{\rm IR} > 11$	240			$3.08^{+1.32}_{-0.81}$	0.07	$1936_{-504}^{+658}$	45	137
ad	$\log L_{\rm IR} < 11$	88			$3.52_{-0.84}^{+0.95}$	0.10	$1718^{+502}_{-339}$	44	122
Xa	$\log L_{\rm IR} > 11$	97	$1.61^{+0.39}_{-0.31}$	0.04	$2.57^{+0.71}_{-0.62}$	0.08			
Xa	$\log L_{\rm IR} < 11 + C I^{\rm cor}$	24	$1.30^{+0.2}_{-0.23}$	0.05	$1.88^{+0.41}_{-0.34}$	0.10			
Xa	$\log L_{\rm IR} < 11$	12	$1.37\substack{+0.34 \\ -0.31}$	0.09	$2.11\substack{+0.18 \\ -0.57}$	0.15			

*Notes.* Means of the optimal conversion parameters  $(X_{CI}, \alpha_{CO}, \kappa_H)$  and the error on the mean  $(\sigma_{\bar{X}}, \sigma_{\bar{\alpha}}, \sigma_{\bar{\kappa}})$  for each subset. We calculate the log-mean and express here in the linear form. We also report the gas-to-dust ratio,  $\delta_{GDR}$ , for a fiducial  $\kappa_{850} = 0.071 \text{ m}^2 \text{ kg}^{-1}$ . We use two normalizations: where dust is one of the tracers, we use  $\kappa^N = 1884 \text{ kg m}^{-2}$  (equivalent to Milky Way  $\delta_{GDR} = 135$  for  $\kappa_{850} = 0.071 \text{ m}^2 \text{ kg}^{-1}$ ); otherwise, for the Xa sample, we use  $X_{CI}^N = 1.6 \times 10^{-5}$  – the mid-range of the values found by Heintz & Watson (2020) for solar metallicity. The errors are the 16th and 84th percentiles of the distribution. The CI<sup>cor</sup> and lo-VALES galaxies are removed for analysis and the variances are derived from the same set.

(excluding He). The sample means,  $\kappa^{\rm N}$  (assumed) and  $\langle \alpha \rangle$  (derived), are next used to estimate an initial gas mass for our galaxy in each of the two tracers,  $L_{\rm 850}$  and  $L'_{\rm CO}$ 

$$M_{\kappa} = \kappa^{N} L_{850} / 4\pi B(\nu_{850}, T_{\rm mw}),$$
  

$$M_{\alpha} = \langle \alpha \rangle L'_{\rm CO}.$$
(18)

Next, we calculate the effective standard deviation by adding the observational error on the tracer luminosities in quadrature to the intrinsic scatter for  $\alpha$  and  $\kappa$ .

$$\sigma_{\text{eff}}^{\kappa} = \sqrt{s_{\kappa}^2 + \sigma_{850}^2},$$
  

$$\sigma_{\text{eff}}^{\alpha} = \sqrt{s_{\alpha}^2 + \sigma_{CO}^2},$$
(19)

where  $\sigma_{850}$ ,  $\sigma_{CO}$  are the errors on log ( $L_{850}$ ) and log( $L'_{CO}$ ), and  $s_{\kappa}$  and  $s_{\alpha}$  are the intrinsic scatter on log( $\kappa_{\rm H}$ ) and log( $\alpha_{\rm CO}$ ) listed in Table 3.

The optimal H<sub>2</sub> mass estimate is then calculated thus

$$M^{\text{opt}} = \frac{M_{\kappa}/\sigma_{\kappa,\text{eff}}^2 + M_{\alpha}/\sigma_{\alpha,\text{eff}}^2}{1/\sigma_{\kappa,\text{eff}}^2 + 1/\sigma_{\alpha,\text{eff}}^2}.$$
(20)

We now work back to find the optimal conversion parameters for this galaxy

$$\kappa_{\rm H}^{\rm opt} = \kappa^{\rm N} M^{\rm opt} / M_{\kappa},$$
  

$$\alpha_{\rm CO}^{\rm opt} = \langle \alpha \rangle M^{\rm opt} / M_{\alpha}.$$
(21)

#### 5.1.2 Single band: empirical conversion factors

The three empirical conversion factors,  $\alpha_{850}$ ,  $\alpha_{CI}$ , and  $\alpha_{CO}$ , directly relate the observable tracer luminosity to a gas mass, according to equation (1). If only one tracer ( $L'_{CO}$ ,  $L'_{CI}$ , or  $L_{850}$ ) is available, the empirical conversion factor we have estimated in Table 7 is the best choice. We adopt a convention that the empirical parameters are referenced to  $M_{mol}$ , which includes a factor 1.36 for He.

Figs 6–9 show the empirical conversion factors as a function of  $L_{IR}$ ; Fig. 10 shows the empirical conversion factors as a function of the tracer luminosity, and Fig. 11 shows their distribution when the sample is split into MS galaxies and SMGs (see Table 8 for details). All empirical factors show significant but shallow correlations with the tracer luminosity. We have carefully accounted for the co-variance

between the x and y parameters when fitting, so the correlations we find are not caused by the involvement<sup>24</sup> of  $L_{850}$ ,  $L'_{CI}$ , and  $L'_{CO}$  in the derivation of  $\alpha_{850}$ ,  $\alpha_{CI}$ , and  $\alpha_{CO}$ . Table 9 lists the fit parameters. The intrinsic scatter in all of these relationships is very small once the measurement errors are accounted for. Correlations are also seen between  $L_{IR}$  and  $\alpha_{850}$ ,  $\alpha_{CI}$ , and  $\alpha_{CO}$  (Fig. 7–9), albeit with more scatter. A correlation between  $\alpha_{850}$  and  $L_{850}$  was also noted by Sco16

A contraining between  $\alpha_{850}$  and  $L_{850}$  was also noted by Scoro (shown as the red dashed line on our plot) but ours is somewhat shallower (m = 0.052 versus m = 0.07 from Sco16), although the difference is unlikely<sup>25</sup> to be significant. These shallow but significant relationships with their tracer luminosity could be further applied to give a more accurate calibration (see Table 9). The trends of  $\alpha_{CI}$  and  $\alpha_{CO}$  with their respective tracers are explored for the first time in large numbers here.

#### 5.2 Discussion of empirical factors relative to the literature.

#### 5.2.1 Submillimetre dust empirical calibration, $\alpha_{850}$

The final calibration factors from this work are provided in Table 10. A compilation of  $\alpha_{850}$  values from our optimal method<sup>26</sup> and those from the literature, referenced to a common  $\alpha_{CO} = 4.3$ : the Galactic value including He from Bolatto et al. (2013) is presented in Table 11. Literature values cover the range,  $\alpha_{850} = 3.6-10.1 \times 10^{12}$ , comfortably within the range of estimates here:  $\alpha_{850}^{\text{opt}} = 6.5-7.2 \times 10^{12}$ . The lowest value,  $\alpha_{850} = 3.6^{+3.6}_{-1.9} \times 10^{12}$ , comes from the local sample of Orellana et al. (2017), who include H I as well as  $M_{\text{mol}}$  (from  $L'_{CO}$ ). Their  $\alpha_{850}$  refers to the total gas mass – sensibly, since their lower luminosity sample is more H I-dominated than the others we compare to – meaning that a lower value for  $\alpha_{850}$  is required. The

 $<sup>^{24}</sup>$ The inclusion of the co-variance matrix in the fit reduces the slope (closer to zero) by 0.05.

 $<sup>^{25}</sup>$ Sco16 do not quote an error on their fit, so it is difficult to be certain, but the error on Sco16 would likely be larger than ours, which means they are consistent to within  $2\sigma$ .

<sup>&</sup>lt;sup>26</sup>There is no significant difference if we fix  $\alpha_{CO}$  to 4.3.



**Figure 6.** Top row: Distributions of the three conversion factors,  $X_{CI}$ ,  $\alpha_{CO}$ , and  $\kappa_{H}$  for the daX sample. The overall normalization has been set as  $\kappa_{H}^{N} = 1884 \text{ kg m}^{-2}$ , which produces the  $\delta_{GDR} = 135$  of the Milky Way for  $\kappa_{850} = 0.071 \text{ m}^2 \text{ kg}^{-1}$  (Jones 2018). The intrinsic pairwise scatter and conversion factors were estimated excluding C I<sup>cor</sup> and lo-VALES galaxies. Lower left-hand panel: the optimized conversion factors as a function of  $L_{IR}$ . The grey shaded band represents  $X_{CI}$  at solar metallicity using the relationship found in GRB and QSO absorber hosts by Heintz & Watson (2020), a measure which is independent of assumptions about  $\alpha_{CO}$  or  $\delta_{GDR}$ . Lower right-hand panel: the running means of the conversion factors, with error bars of  $\sigma/\sqrt{N_{bin}}$ . The solid shaded bins are the means for the grey points which are those used to determine the calibration, the yellow points are C I<sup>cor</sup> and the semitransparent pentagon is the mean of those – see Section 4.4 for more details.

highest value,  $\alpha_{850} = (10.1 \pm 0.3) \times 10^{12}$ , is from Sco16.<sup>27</sup> There are two reasons why Sco16 found a significantly higher  $\alpha_{850}$  compared

to our analysis. The first is simple mathematics, as Sco16 quote a linear mean for a distribution that has a significant tail to higher

<sup>27</sup>The original value of  $\alpha_{850} = 6.7 \times 10^{12}$  quoted by Sco16 assumed that  $\alpha_{CO} = 6.5$  to calibrate the gas mass from CO. Re-normalizing the Sco16

result to the same  $\alpha_{CO} = 4.3$  as our literature comparison increases the Sco16 value to  $\alpha_{850}(\alpha_{CO} = 4.3) = 10.1 \times 10^{12}$ .



**Figure 7.** Top panel: Distributions of  $X_{CI}$  and  $\kappa_{H}$  for the Xd sample, where the normalization used is  $\kappa_{H}^{N} = 1884 \text{ kg m}^{-2}$ . Left-hand panel: optimized conversion factors as a function of  $L_{IR}$  for the galaxies in the Xd sample. The grey shaded bands represent the range of  $X_{CI}$  ( $Z = Z_{\odot}$ ) from a study of GRB and QSO absorber hosts by Heintz & Watson (2020), a measure which is independent of assumptions about  $\alpha_{CO}$  or  $\delta_{GDR}$ , and also the range of  $\kappa_{H}$  in nearby galaxies. Right-hand panel: running means of the conversion factors with error bars of  $\sigma/\sqrt{N_{bin}}$ . The solid coloured pentagons are the means for the grey points, which are those used to determine the conversion; the yellow points are the C I<sup>cor</sup> galaxies and the semitransparent pentagon is the mean of those (see Section 4.4 and Appendix B2). The grey dashed error bars in the  $X_{CI}$  running mean are for the so-called simple method, where  $\kappa_{H} = 1884 \text{ kg m}^{-2}$  is fixed for all galaxies.

values; in contrast, we quote a log-mean which is less sensitive to tails. This statistical bias results in a linear mean for  $\alpha_{850}$  which is 20 per cent higher than the log-mean (Behroozi, Wechsler & Conroy 2013). Assuming the shape of our  $\alpha_{850}$  distribution is similar to that from Sco16, we adjust their linear mean down by 20 per cent to approximate our log-mean method. Thus, our log-mean estimate of

the Sco16 value is  $\alpha_{850}^{\text{LM}}(\text{Sco16}) = 8.4 \times 10^{12}$ . Secondly, there have been changes in the versions of the *Herschel* pipeline data used as the basis for the local portion of the Sco16 sample (see Appendix A). When we re-fitted the local galaxy SEDs to estimate  $T_{\text{mw}}$  and  $L_{850}$ using the most recent *Herschel* flux densities (Chu et al. 2017; Clark et al. 2018), we found an increase in  $L_{850}$  of ~0.1 dex compared to



Figure 8. Top panel: Distributions of  $X_{CI}$  and  $\alpha_{CO}$  for the Xa sample, where the normalization has been set to  $X_{CI}^N = 1.6 \times 10^{-5}$  – the middle of the range for  $Z_{\odot}$  found in GRB hosts and QSO absorbers by Heintz & Watson (2020). Lower left-hand panel: optimized conversion factors as a function of  $L_{IR}$  for the galaxies in the Xa sample. Right-hand panel: running means of the conversion factors with error bars of  $\sigma/\sqrt{N_{bin}}$ . The solid coloured pentagons are the means for the grey points which are those used to determine the conversion; the yellow points represent the C1<sup>cor</sup> galaxies and the semitransparent pentagon is the mean of those – see Section 4.4 and Appendix B2. The grey dashed error bars in the  $\alpha_{CO}$  running mean are for the so-called simple method, where  $X_{CI} = 1.6 \times 10^{-5}$  is fixed for all galaxies.

that reported in Sco16.<sup>28</sup> Once these factors are accounted for, the Sco16 result is comparable to ours.

<sup>28</sup>This difference is not just the photometry change; Sco16 used a different

method to estimate  $L_{850}$  from the Herschel 500- $\mu$ m fluxes.

#### Atomic carbon empirical factor: $\alpha_{CI}$

We compare our optimized  $\alpha_{CI}$  estimates with others from the literature in Table 12, and for reference, we also compile the literature values for  $X_{CI}$  in Table 13, the two are related simply by the excitation factor  $Q_{10}$ , as given in equation (11). Our values are a



Figure 9. Top panel: Distributions of  $\alpha_{CO}$  and  $\delta_{GDR}$  for the ad sample, where the normalization has been set to  $\kappa_{H}^{N} = 1884 \text{ kg m}^{-2}$ . Lower left-hand panel: optimized conversion factors as a function of  $L_{IR}$  for the galaxies in the ad sample. The cyan diamonds indicate galaxies with  $f_{HI} > 1$  which have been corrected for the contribution of dust mixed with H I, following the procedure outlined in Appendix B1. The open peach circles are the lo-VALES galaxies which we suspect have  $f_{HI} > 1$  – these are not included in the analysis. Right-hand panel: running means of the conversion factors with error bars of  $\sigma/\sqrt{N_{bin}}$ . The coloured points reflect the optimized results and the grey dashed error bars for the  $\alpha_{CO}$  running mean are for the so-called simple method, where  $\kappa_{H} = 1884 \text{ kg m}^{-2}$  is fixed for all galaxies.

weighted average of the three samples that contain CI, where we find  $\langle \alpha_{CI} \rangle = 17.3 \pm 0.3$  (standard error on the mean). This compares well with the only truly independent measure,  $\alpha_{CI} = 21.4^{+13}_{-8}$ , from absorber systems across a range of redshift by Heintz & Watson (2020), but is considerably higher than reported by many literature studies, e.g.  $\alpha_{CI} = 4.9$ –10.3 for a large study of (U)LIRGs by Jiao

et al. 2017 and  $\alpha_{\rm CI} = 7.3^{+6.9}_{-3.6}$  for local discs by Crocker et al. (2019). Many literature studies assume a fixed value for either  $X_{\rm CI}$  or  $\alpha_{\rm CO}$  in order to derive  $\alpha_{\rm CI}$  (typically  $X_{\rm CI} = 3 \times 10^{-5}$  or  $\alpha_{\rm CO} = 1$  for high-*z* SMG or local (U)LIRGs). These assumed values are very different from those we have derived here under our minimal assumption that metal rich galaxies have similar dust prop-

 Table 7. Empirical conversion factors recommended for use when only a single gas tracer observation is in hand. These include a factor 1.36 to account for He.

Sample	$lpha_{850}( imes 10^{12})$ (W Hz <sup>-1</sup> M <sub>☉</sub> <sup>-1</sup> )	$lpha_{ m CO}$ ${ m M}_{\odot}$ (K km	$\frac{\alpha_{\rm CI}}{\rm s^{-1}pc^2)^{-1}}$
$\log L_{\rm IR} < 11$	$5.8 \pm 0.1$	$4.7 \pm 0.1$	_
$\log L_{\rm IR} > 11$	$6.9 \pm 0.1$	$4.0 \pm 0.1$	$17.0\pm0.3$
MS	$6.2 \pm 0.1$	$4.4 \pm 0.1$	$19.1\pm0.6$
SMGs	$7.3 \pm 0.1$	$3.8\pm0.1$	$16.2\pm0.4$

*Notes.* Values are the weighted means and errors from each of the daX, ad, and dX samples which all have the same normalization of  $\kappa_{\rm H}^{\rm N} = 1884 \, {\rm kg \, m^{-2}}$ . Low luminosity (log  $L_{\rm IR} < 11$ ) values are based only on the ad sample due to the small numbers of reliable C I measurements in this luminosity range. The differences in the weighted means for SMGs and MS galaxies are significant in all cases, but are likely driven by the trends with luminosity seen in Figs 6, 9, and 10. A more accurate way to determine the conversion factor to use would be to use one of the relationships from Table 9.

erties. Table 12 describes the assumptions made for each literature source.

The MS galaxies in this work have a value of  $\alpha_{CI}$  of 19.1  $\pm$  0.6, again significantly higher than that found in Crocker et al. (2019). However, J19, using a largely overlapping sample, derived a value of  $\alpha_{CI} = 19.9 \pm 1.9$  using the same *Herschel* FTS CI mapping observations. Crocker et al. use  $L'_{[CO](2-1)}$  images and spatially resolved  $\alpha_{CO}$  estimates from Sandstrom et al. (2013), which were derived using a robust method which minimizes the scatter in the gas-to-dust ratio (see also Eales et al. 2012). While there are no a priori assumptions about  $\alpha_{CO}$  in Crocker et al. (2019), implicit assumptions are required for the average CO  $r_{21}$  excitation. The limited sensitivity of the FTS instrument to [CI](1-0) meant that CI was primarily detected in the brighter nuclear regions, where  $\alpha_{CO}$ tends to be lower than is typical in spiral discs ( $\sim$ 1 compared to  $\sim$ 3– 4), (e.g. Sandstrom et al. 2013) - as noted by Crocker et al. Their measured  $L'_{CI}/L'_{CO}$  ratios in the resolved regions are compatible with other MS galaxies in our sample (though still higher than the ratios derived for the same set of sources by J19, see Fig. 5), meaning that the  $\alpha_{\rm CI}/\alpha_{\rm CO}$  ratios are also similar. As  $\alpha_{\rm CO}$  in these regions is determined to be low in the Sandstrom et al. (2013) analysis, the  $\alpha_{CI}$ inferred by Crocker et al. is correspondingly lower as well.

Jiao et al. (2021; hereafter J21) use CO(1–0), H I, [C I](1–0), dust continuum, and metallicity maps to investigate the variation of  $\alpha_{CI}$ and  $\alpha_{CO}$  across the discs of six well-resolved local galaxies from their J19 study. They use the FIR/submm dust maps from Spitzer and Herschel and the method of Draine et al. (2007) to model the dust mass across the galaxy and relate this to a gas mass via a relationship between dust-to-gas and metallicity (Draine et al. 2007; Muñoz-Mateos et al. 2009; Sandstrom et al. 2013). As they explicitly use the dust mass together with a model of the  $\delta_{GDR}$  dependence on metallicity, their results are normalized to the dust properties of the Draine & Li (2007; hereafter DL07) model, which assumes  $\delta_{\rm GDR} = 100$  and  $\kappa_{850} = 0.034 \,\mathrm{m}^2 \,\mathrm{kg}^{-1}$  at solar metallicity. Their self-consistent DGR(ii) method derives weighted mean values of  $\langle \alpha_{\rm CI} \rangle = 19.9 \pm 1.9$  (including the information from lower limits) and  $\langle \alpha_{\rm CO} \rangle = 2.0 \pm 0.3 \ (2.6 \pm 0.4)$  over the same area as the C I observations (the entire CO detection region).<sup>29</sup> In the central region, the  $\alpha_{\rm CO}$  values are significantly lower at  $\alpha_{\rm CO}^{\rm C} = 1.5 \pm 0.3$ , while  $\alpha_{\rm CI}$ is not found to be significantly different with  $\alpha_{CI}^{C} = 21.8 \pm 0.5$ .

These values are comparable to our average of  $\alpha_{\rm CI} = 19.1 \pm 0.6$  for MS galaxies, and to the average value for  $Z_{\odot}$  derived independently by Heintz & Watson (2020) of  $\alpha_{\rm CI}$ (HW20) =  $21.4^{+13.3}_{-8.2}$ .<sup>30</sup>

## CO empirical factor: $\alpha_{CO}$

We compare our optimized  $\alpha_{CO}$  estimates with others from the literature in Table 14. Sophisticated LVG modelling with very large data sets which include high-density gas tracers, optically thin CO isotopologues, full CO SLEDs, and sometimes the C I lines and dust emission (e.g. Weiß et al. 2007; Papadopoulos et al. 2012b, 2014; Israel 2020; Harrington et al. 2021) can break some of the model degeneracies of the optically thick CO lines, though the method is still reliant on assumptions for [CO/H<sub>2</sub>], isotopologue ratios, the number of components allowed (single components give very different results to multiple components) and the allowed range of velocity gradients in the models.

The best examples are NGC 6240 (Papadopoulos et al. 2014) and the *Planck* lensed galaxies (Harrington et al. 2021) where detailed LVG modelling and comprehensive data sets have sufficient constraints to break the degeneracies which usually be-devil this method. The two-component LVG result for NGC 6240 is  $\alpha_{CO} = 2$ -4 (Papadopoulos et al. 2014; cf.  $\alpha_{CO} = 0.6$  when using a single component LVG model Papadopoulos et al. 2012b) and we can further use the ratio of  $L'_{CI}/L'_{CO}$  measured by Cicone et al. (2018) and our relationship,  $L'_{\rm CI}/L'_{\rm CO} = \alpha_{\rm CO}/\alpha_{\rm CI} = 3324 \,\alpha_{\rm CO} X_{\rm CI}$ , to infer that  $X_{\rm CI} = 1.4-2.9 \times 10^{-5}$  in the starburst region. In fact, our optimized values for this galaxy using global fluxes, are  $\alpha_{CO}(daX) =$  $2.9 \pm 0.6, X_{CI}(daX) = (2.4 \pm 0.5) \times 10^{-5}, \kappa_{H}(daX) = 2800 \pm 700$  $(\delta_{GDR} = 200)$ , in excellent agreement. The *Planck* lensed galaxies analysed by Harrington et al. (2021) do not have the same degeneracy-breaking lines used by Papadopoulos et al. (2014) in their analysis, but they do have multi-J CO coverage and incorporate the CI lines and the dust continuum emission in their model fitting, based on Weiß et al. (2007). They assume similar dust parameters as we do for their normalizations ( $\delta_{GDR} = 120-150$  with  $\kappa_{850} =$ 0.08 m<sup>2</sup> kg<sup>-1</sup>). With this, they infer an average  $\alpha_{CO} = 3-4$  and an average  $\alpha_{CI} = 16.2 \pm 7.9$  (including He), remarkably consistent with our results, given our very simple approach.

## 5.3 Lack of bi-modality in the conversion factors

Our sample contains normal star-forming galaxies – those obeying the SFR– $M_{\star}$  correlation that forms as a result of the more intimate relationship between SFR and H<sub>2</sub> – as well as many extreme star-forming systems, which belong to the (U)LIRG and high-*z* submillimeter selected samples. Here, we remind the reader that we refer to the extreme SF group – those that supposedly require a lower  $\alpha_{CO}$  – as 'SMGs', and the normal star forming sources as 'MS galaxies', or sometimes just 'MS'. As mentioned in Section 2, the assignment of the galaxies to either category is by nature of the data

<sup>30</sup>While there are differences in the normalization for the dust mass model chosen by J21 and ourselves, the introduction of a metallicity dependence for the dust-to-gas ratio by J21 means that there is no simple way to scale their results to our method. However, we can calculate their average 'effective  $\kappa_{\rm H}$ ',  $\kappa_{\rm eff} = 1300$ , which indicates that J21 derive a lower H<sub>2</sub> for a given  $L_{850}$  compared to our normalization (and hence a lower value of  $\alpha_{\rm C1}$  and  $\alpha_{\rm C0}$ ). However, the six galaxies in J21 are a subset of the C I<sup>cor</sup> objects, at the low luminosity end where there are potential decreases in  $\kappa_{\rm H}$  and increases in  $\alpha_{\rm C1}$ .

 $<sup>^{29}\</sup>mbox{We}$  have multiplied the values in J21 by 1.36 to include He for consistency with our convention.

Table 8.	Log means and	statistical tes	sts for conversion	factors for MS	galaxies and SMGs.
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Ст	Sample	$X_{\rm CI}({ m MS})$	$X_{\rm CI}({ m SMG})$	$\alpha_{\rm CI}({\rm MS})$	$\alpha_{\rm CI}({\rm SMG})$	$Z(\sigma)$	$P_{\rm KS}$	Р <sub>КS</sub> (25 к)
	daX	$1.41 \pm 0.07$	$1.70 \pm 0.06$	$19.1 \pm 0.9$	$15.8 \pm 0.5$	3.1	0.016	0.074
	Xd	$1.44 \pm 0.05$	$1.65\pm0.05$	$18.7 \pm 0.8$	$16.4 \pm 0.5$	2.6	0.013	0.48
	Xa	$1.44 \pm 0.04$	$1.64\pm0.05$	$18.7\pm0.6$	$16.4\pm0.5$	3.0	0.028	
CO		$\alpha_{\rm CO}({\rm MS})$	$\alpha_{\rm CO}({\rm SMG})$					
	daX	$2.56\pm0.14$	$2.61\pm0.12$			0.3	0.35	0.017
	Xa	$2.45\pm0.12$	$2.70\pm0.10$			1.6	0.05	
	ad	$3.32\pm0.07$	$2.87 \pm 0.09$			3.8	0.0008	0.04
Dust		$\kappa_{\rm H}({\rm MS})$	$\kappa_{\rm H}({\rm SMG})$	$\alpha_{850}(MS)$	$\alpha_{850}(SMG)$			
	daX	$1758 \pm 90$	$2039 \pm 113$	$6.8 \pm 0.3$	$7.7 \pm 0.3$	2.2	0.04 (0.25)	0.76
	Xd	$1757 \pm 66$	$2025 \pm 58$	$6.9 \pm 0.2$	$7.4 \pm 0.2$	3.0	0.015 (0.41)	0.58
	ad	$1722 \pm 36$	1981 ± 65	$6.0 \pm 0.1$	$7.3 \pm 0.2$	3.6 (6.5)	$\begin{array}{c} 0.0017 \\ (4 \times 10^{-11}) \end{array}$	0.11

*Notes.* We compare MS galaxies and SMGs for each subset to look for differences in the parameters. The MS group excludes C 1<sup>cor</sup> and lo-VALES galaxies, but it does include the  $f_{\rm HI}$ >1 galaxies, after applying the correction from Appendix B1. The numbers in each subset are: daX: MS = 46, SMG = 55; Xd: MS = 60, SMG = 79; Xa: MS = 54, SMG = 55; ad: MS = 184, SMG = 144. Bold indicates parameters which are significantly different between the MS galaxies and SMGs in the Z-test and KS tests. The C I parameters,  $X_{\rm CI}$  and  $\alpha_{\rm CI}$ , are simply linked due to our adoption of constant  $Q_{10} = 0.48$ , meaning that the distributions have the same KS results. The dust parameters,  $\kappa_{\rm H}$  and  $\alpha_{850}$ , are related to each other as a function of  $T_{\rm mw}$  and so they can behave differently, e.g.  $\kappa_{\rm H}$  can be indistinguishable between samples but the  $\alpha_{850}$  can be significantly different. Thus in the dust section, there are two  $P_{\rm KS}$  values: those for  $\kappa_{\rm H}$  and then, in parentheses, those for  $\alpha_{850}$ . The final column,  $P_{\rm KS}(25 \, \text{K})$ , is the KS result when  $T_{\rm mw}$  is fixed to 25 K, which makes the distributions of  $\kappa_{\rm H}$  and  $\alpha_{850}$  identical.

rather 'fuzzy' as we do not have a measure of SFR or stellar mass for all sources, nor any homogeneous way to estimate them. We, thus, rely on the categories used by previous authors where possible, especially for high-z sources. The z = 1 galaxies from the samples of Bourne et al. (2019) and Valentino et al. (2018, 2020) are deemed to be 'MS', as are the sources from ASPECs (Boogaard et al. 2020). Most low redshift sources with log  $L_{IR} < 12$  are classed as 'MS' though there are some exceptional LIRG class sources in the local Universe which have extreme properties as evidenced by their FIR, MIR lines, and vibrational HCN (Díaz-Santos et al. 2017; Falstad et al. 2021). We note that using a more conservative separation when assigning galaxies into MS and extreme starburst categories does not change any of the results. We therefore conclude that while our assignment of sources into the two SF categories is not perfect, this categorization is not capable of masking any strong bi-modality in the observable ratios.

Fig. 11 and Table 8 detail the distributions of conversion factors for each sample, split into MS galaxies and SMGs. While formally there are significant differences in the parameters for some samples, these are very small – around 10–20 per cent in the mean, rather than the factor ~3–4 times often assumed for  $\alpha_{CO}$  (e.g. Downes & Solomon 1998,  $\alpha_{CO} = 0.8$ , derived for four ULIRGs). In fact, only the ad sample shows any difference in  $\alpha_{CO}$  between the MS galaxies and SMGs, while the estimates based on C1 and CO, or on all three tracers, show no significant difference. This is partially explained by the larger luminosity range in the ad sample, combined with the previously noted negative correlation between  $\alpha_{CO}$  and luminosity (Figs 9 and 10), with a factor ~2 times reduction in  $\alpha_{CO}$  for a factor ~100 times increase in  $L'_{CO}$ . We cannot rule out that the correlation of  $\alpha_{CO}$  with luminosity is the true reason that the ad sample shows a significant difference MS galaxies and SMGs.<sup>31</sup>

 ${}^{31}L_{\rm IR}({\rm ad})({\rm MS}) = 10.95$  ( $L'_{\rm CO}({\rm ad})({\rm MS}) = 9.31$ ) while  $L_{\rm IR}({\rm Xa})({\rm MS}) = 11.22$  ( $L'_{\rm CO}({\rm Xa})({\rm MS}) = 9.52$ ). Using the relation in Table 9, the expected  $\Delta\alpha_{\rm CO} = \alpha_{\rm CO}({\rm MS}) - \alpha_{\rm CO}({\rm SMG}) = 0.36$  for the Xa sample and – due to the lower numbers in the Xa and daX samples – such a difference would not be detected at a significant level, if it existed.

This is not the first time<sup>32</sup> that lack of bi-modality in  $\alpha_{CO}$  has been reported when compared to dust-based determinations (e.g. Magdis et al. 2012; Rowlands et al. 2014; Genzel et al. 2015). The range of  $\alpha_{CO}$  we find for SMGs (see Fig. 9) is well within the framework set out by Papadopoulos et al. (2012a), who noted that galaxies with a highly turbulent ISM (e.g. ULIRGs and SMGs) can have  $\alpha_{CO}$  similar to galaxies with a much more quiescent ISM, the only difference being that in a turbulent ISM, the distribution of gas mass as a function of density is weighted to higher densities than in a less-turbulent ISM.

Recent joint SLED/SED modelling of an exquisite data set that includes CO, CI, and dust continuum for lensed SMGs (Harrington et al. 2021) finds a mean  $\alpha_{CO} = 3.4$ -4.2 for these highly turbulent galaxies (albeit with a large dispersion). The Harrington et al. radiative transfer models employ a continuum distribution of molecular gas mass as a function of average Mach number (and average density of the molecular cloud ensemble), making them better equipped to 'capture' any re-distribution of the underlying molecular gas mass towards higher densities. While important for other issues (e.g. the initial conditions of star formation in SMGs/ULIRGs), such a re-distribution in a highly turbulent ISM may actually leave  $\alpha_{CO}$ statistically unaffected. The initial reports of a bimodal  $\alpha_{CO}$  factor in the local Universe, with  $\sim$ 4–5 times lower values for ULIRGs than LIRGs and ordinary spirals, can possibly be explained by a COluminous, strongly unbound, low-density molecular gas component found preferentially in ULIRGs. Such a component can dominate the global CO(1-0) line luminosities of ULIRGs/SMGs (even if containing only small fractions of their total molecular gas), while its large  $K_{vir}$  values will yield systematically low  $\alpha_{CO}$  factors, under one-component LVG modelling (equation 9).<sup>33</sup>

<sup>&</sup>lt;sup>32</sup>However, our current data set is more homogeneous, using only CO(1–0) or CO(2–1) and a consistent approach to modelling the dust with our empirical relations for  $T_{\rm mw}$ .

 $<sup>^{33}</sup>$ Also, we must consider the size of the original sample – four ULIRGs in the first study by Downes & Solomon (1998).



**Figure 10.** Correlations of the empirical calibration factors with their tracer luminosity. The largest sample was used in each case: ad (top and bottom); Xd (top centre). For  $\alpha_{850}$  (top), we include the relationship from Scoville et al. (2016), exactly as quoted in that paper. Parameters for the median posterior fit (dark blue line), accounting for co-variance in the errors, are quoted in the legend, and we include 100 random fits from sampling the posterior as pale blue lines to show the scatter. Further details are given in Table 9. All tracers have a significant correlation with their tracer luminosity.

For individual galaxies, only multicomponent models of SLED/SED (that also include molecules/transitions tracing the dense gas) can properly account for this effect (e.g. Papadopoulos et al. 2014; Harrington et al. 2021), while for large galaxy samples, our cross-calibration of  $\alpha_{CO}$  against the other two molecular gas mass tracers, is the most economical method. In that regard, it is worth noting that *dust continuum is immune to the gas-dynamics effects described above*, i.e. a diffuse low-density, unbound, H<sub>2</sub> gas component will contribute very little to the total dust continuum if its gas/dust mass is indeed low. The optically thin C I line emission will also be much less sensitive than CO(1–0) to such gas-dynamics effects exactly because of its low optical depths. These are perhaps the reasons why our cross-calibration of  $\alpha_{CO}$  against dust and C I emission has not uncovered any obvious bimodality of its values in MS galaxies compared to SMGs.

The range of values we find for  $\alpha_{\rm CO}$  is consistent with expected values for  $Z > 0.5 Z_{\odot}$  galaxies (Accurso et al. 2017, based on calibrating  $\alpha_{\rm CO}$  using C II). Using their predictions, we would expect  $2.7 < \alpha_{\rm CO} < 15.2$  for the likely range of metallicity and offset from the MS in our sample.

Our underlying assumption: that the dust–gas properties of MS galaxies and SMGs can be described as a uni-modal distribution with well-defined mean and scatter, is based on our finding that the luminosity ratios (Fig. 2) – the most basic observables used in deriving the empirical conversion factors – have such a distribution. They show no evidence for the strong bi-modality advocated for  $\alpha_{\rm CO}$  in some of the literature. That the distribution of the observed luminosity ratio is, to first order, similar to the distribution of the conversion parameters, is the simplest 'Occam's Razor' assumption we can make.

To see what a different initial assumption would mean for the conversion factors, we repeated our analysis, this time inserting the popular bi-modal behaviour in  $\alpha_{CO}$  (Greve et al. 2005; Weiß et al. 2005; Tacconi et al. 2006, 2008; Genzel et al. 2010; Walter et al. 2011; Alaghband-Zadeh et al. 2013; Jiao et al. 2017; Valentino et al. 2018) as our prior, such that the sample mean normalizations for SMGs and MS galaxies are set to be different:  $\alpha_{CO}^{N}(SMG) = 0.8$  and  $\kappa_{H}^{N}(MS) = 1884 \text{ kg m}^{-2}$ . Our optimal method then allows the data to return the most likely values for the other parameters<sup>34</sup> under these assumptions.

Fig. 12 shows the results with this bi-modal normalization, (blue points: MS galaxies, red points: SMGs). By design, we have reproduced the extreme bi-modality  $\alpha_{CO}(MS) \sim 3-4$  and  $\alpha_{CO}(SMG) \sim 1$ , but Fig. 12 clearly shows that the same extreme bi-modality has to be present in  $\kappa_{\rm H}$  ( $\delta_{\rm GDR}$ ) and  $X_{\rm CI}$ , giving a clear prediction that  $X_{\rm CI}({\rm SMG}) \ge 4 \times 10^{-5}$  if the bimodality in  $\alpha_{\rm CO}$  really exists, with essentially no overlap in  $X_{CI}$  between the MS galaxies and SMGs. To test this will require an independent determination of  $X_{CI}$  in SMGs, without reference to dust or CO calibration. To date, there is no such determination of  $X_{CI}$ , although Izumi et al. (2020) observed the nearby LIRG NGC 7469 with ALMA, using kinematic data to derive  $M_{dyn}$ , which is the sum of  $M_{H2}$ , stellar mass, and dark matter. This method has promise, but the systematic uncertainties in  $M_{\rm H2}$  from this analysis are too large (0.3 dex) to answer our question. While the Izumi et al. study clearly indicates<sup>35</sup> that the  $[C^0/CO]$  abundance can be enhanced in extreme environments, the CND is only a tiny region and the global ratio for this source is very similar to other

<sup>&</sup>lt;sup>34</sup>We did not re-calculate the intrinsic scatter split into MS galaxies and SMGs, only the values of the  $X_{CI}/\alpha_{CO}$  and  $\alpha_{CI}/\kappa_{H}$  pairs.

<sup>&</sup>lt;sup>35</sup>Via the extremely high observable ratio  $L'_{\rm CI}/L'_{\rm CO} = 0.92$  in the CND.



**Figure 11.** The empirical conversion factors,  $\alpha_{CO}$ ,  $\alpha_{CI}$ , and  $\alpha_{850}$ , separately for MS galaxies and SMGs. The blue line histograms represent the MS galaxies. The dark blue dashed lines show the  $f_{HI} > 1$  and C I<sup>cor</sup> galaxies. We keep these separate because the corrections are uncertain and the C I<sup>cor</sup> galaxies are significantly different in their distributions to the rest of the MS galaxies, as discussed in Section 4.4. The SMG histograms are shown in red. The result of two-sample KS tests are shown on each panel for each of the comparisons (MS galaxies versus SMGs). The main comparison between MS galaxies and SMGs is shown first; below, in parentheses, are the results for comparison between the robust MS galaxies and those requiring the corrections described above. daX: Only  $\alpha_{CI}$  shows a significant (3.2 $\sigma$ ) difference between the two groups, decreasing to 1.9 $\sigma$  if constant  $T_{mw} = 25$  K is used. Xa: There is a marginal difference in  $\alpha_{C1}$  (2.5–2.9 $\sigma$ ) but no difference in  $\alpha_{C0}$ . There is a significant difference between the C I<sup>cor</sup> and other MS galaxies (dotted blue line and parentheses), which cannot be attributed to uncertain aperture corrections as CO and C I are measured in the same aperture. Xd: For  $\alpha_{CI}$  (and  $\kappa_{H}$  – see Table 8), there is a marginal difference (2.6 $\sigma$ -3.0 $\sigma$ ) between the MS galaxies and SMGs, where these differences vanish when using fixed  $T_{mw} = 25$  K. There is no difference between MS galaxies and SMGs for  $\alpha_{850}$ . ad: There is a small (~10 per cent) difference in the mean  $\alpha_{C0}$  between the MS galaxies and SMGs, significant at the 3.5 $\sigma$  level. This drops to 2.5 $\sigma$  when comparing only log  $L_{IR} > 11$  MS galaxies, and drops further when using constant  $T_{mw} = 25$  K. We conclude that this is a luminosity-driven effect, not a property of the star-formation mode *per se*. The difference in  $\alpha_{850}$  is highly significant (as is the difference in  $\kappa_{H}$ , to a lesser extent – see Table 8). Fig. 10 shows that this difference is the result o

y	x	т	С	rs	Ν	Sample
$\log \alpha_{\rm CO}$	$\log L'_{\rm CO}$	-0.062 (0.009)	1.25 (0.09)	-0.33	335	ad
$\log \alpha_{850}$	$\log L_{850}$	0.052 (0.008)	11.60 (0.18)	0.37	335	ad
$\log \alpha_{CI}$	$\log L_{\rm CI}'$	-0.052 (0.013)	1.73 (0.12)	-0.3 (0.0003)	140	Xd

**Table 9.** Fits to the conversion parameters and their tracer luminosities. All quantities include He.

*Notes.* Fits in the form y = mx + c for the empirical conversion parameters and their tracer luminosities. The C1<sup>cor</sup> galaxies are excluded from the fits but are shown in the plots (Fig. 10).  $r_s$  is the Spearman rank correlation coefficient with probability of the null hypothesis of no correlation in parentheses for  $p \ge 0.0001$ . The effects of co-variance in the errors have been accounted for.

Table 10. Empirical calibration factors derived from this study.  $M_{mol}$  columns include a factor of 1.36 for He.

		M <sub>H2</sub>			M <sub>mol</sub>			
Sample	Ν	$\alpha_{850} (\times 10^{12})$ W Hz <sup>-1</sup> M <sub>☉</sub> <sup>-1</sup>	$lpha_{ m CO}$ M $_{\odot}$ (K km	$\frac{\alpha_{\rm CI}}{\rm ns^{-1}pc^2)^{-1}}$	$\alpha_{850} (\times 10^{12})$ W Hz <sup>-1</sup> M <sub>☉</sub> <sup>-1</sup>	$lpha_{ m CO}$ M $_{\odot}$ (K km	$\alpha_{CI} \approx 1 \ s^{-1} \ pc^2)^{-1}$	
daX	101	$9.9 \pm 0.2$	$2.6\pm0.1$	$12.6\pm0.4$	$7.3 \pm 0.2$	$3.5\pm0.1$	$17.2\pm0.5$	
ad $(\log L_{\rm IR} > 11)$	240	$9.1 \pm 0.1$	$3.1 \pm 0.1$		$6.7 \pm 0.1$	$4.2 \pm 0.1$		
ad	326	$8.8 \pm 0.1$	$3.2 \pm 0.1$		$6.5 \pm 0.1$	$4.3 \pm 0.1$		
Xd	140	$9.8 \pm 0.3$		$12.7\pm0.4$	$7.2 \pm 0.2$		$17.3\pm0.5$	
Xa	109		$2.5\pm0.1$	$12.5\pm0.3$		$3.4\pm0.1$	$17.0\pm0.4$	

*Notes.* The samples which include dust continuum used a normalization of  $\kappa_{\rm H}^{\rm N} = 1884 \, {\rm kg \, m^{-2}}$ , while for the Xa sample,  $X_{\rm CI}^{\rm N} = 1.6 \times 10^{-5}$  was used. In this analysis, we excluded the C 1<sup>cor</sup> and lo-VALES galaxies (see Section 4.4).

**Table 11.** Summary of our empirical dust continuum– $M_{mol}$  calibration factor,  $\alpha_{850}$ , compared to literature values referenced to  $\alpha_{CO} = 4.3$  (i.e. Galactic  $\alpha_{CO}$ , including He).

$\frac{\alpha_{850}(\times 10^{12})}{(W \text{Hz}^{-1} \text{M}_{\odot}^{-1})}$	Sample	Ngal	Notes	Reference
$6.4 \pm 0.1$	All	328	Log-mean opt	This work
$7.2 \pm 0.2$	SMGs	144	Log-mean opt	This work
$5.9 \pm 0.1$	MS	184	Log-mean opt	This work
$10.1\pm0.3$	Local galaxies and SMGs	72	Linear mean	Scoville et al. (2016)
8.3	MS	30	Linear mean	Scoville et al. (2016)
12.7	SMGs	30	Linear mean	Scoville et al. (2016)
$3.6^{+3.6}_{-1.9}$	Local galaxies	136	Median H I + $4.3L'_{CO}$	Orellana et al. (2017)
$6.1 \pm 0.14$	$z < 0.4$ 160- $\mu$ m selected	41	Log-mean (ex lovales)	Hughes et al. (2017)
$8.4 \pm 1.0$	z = 1.6-2.9 unlensed SMGs <sup>a</sup>	9	Log-mean	Kaasinen et al. (2019)
$11.6\pm1.2$	z = 1.6-2.9 unlensed SMGs <sup>a</sup>	9	Linear mean	Kaasinen et al. (2019)

*Notes.* Errors quoted are the standard error on the mean, from the variance of the  $L_{850}/L'_{CO}$  ratio. Where we have the data for  $L'_{CO}$  and  $L_{850}$ , we calculate the mean log  $\alpha_{850}$  because the distribution of ratios is skewed in linear space (Behroozi et al. 2013), leading to a significantly higher value for  $\alpha_{850}$  in the linear averaging. We also cite the linear average, scaled to  $\alpha_{CO} = 4.3$  where that is presented in the original literature reference. <sup>*a*</sup>This small sample may potentially be biased by choosing the brightest 850- $\mu$ m galaxies from the parent sample.

**Table 12.** Summary of our empirical  $\alpha_{CI}$  calibration compared to work from the literature;  $\alpha_{CI}$  is quoted including He.

$\frac{\alpha_{\rm CI}}{M_{\odot}  ({\rm K  km  s^{-1}  pc^2})^{-1}}$	Sample	$N_{\rm gal}$	Notes	Reference
$17.0 \pm 0.3$	$\text{Log } L_{\text{IR}} > 11$		Weighted average	This work
$19.1\pm0.6$	MS		Weighted average	This work
$16.2 \pm 0.4$	SMGs		Weighted average	This work
$10.3\pm0.3$	(U)LIRGs	71	Assuming $X_{CI} = 3 \times 10^{-5}$	Jiao et al. (2017)
$4.9 \pm 0.3$	(U)LIRGs	71	$CO(1-0)$ with $\alpha_{CO} = 1.1$	Jiao et al. (2017)
$7.3^{+6.9}_{-3.6}$	Resolved local discs	18	CO(2–1) and resolved $\alpha_{CO}$ from S13	Crocker et al. (2019)
$19.9 \pm 1.9$	Resolved local discs	6	H I, CO(1–0), C I, dust, $Z (\kappa_{\rm eff} \sim 1300)$	Jiao et al. (2021)
$16.2\pm7.9$	z = 3 lensed SMGs	16	Multi-J CO, C I dust modelling	Harrington et al. (2021)
$21.4^{+13.3}_{-8.2}$	GRB/QSO absorbers	19	$H_2$ and CI absorption lines at $Z_{\odot}$	Heintz & Watson (2020)
17.6	Theory		For $\zeta_{CR} = 5 \times 10^{-17} s^{-1}$	Offner et al. (2014)

Notes. The values from this work are the weighted averages of the results from each of the three sub-groups containing C1 information.

Table 13.	Summary	of our $X_{\rm C}$	I calibrations	compared	to other	work in th	e literature.
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$\overline{X_{\rm CI}(\times \ 10^{-5})}$	Sample	$N_{\rm gal}$	Notes	Reference
$1.6^{+0.5}_{-0.4}$	$z = 0-5 \log L_{\rm IR} > 11$	90	$L_{850}$ , CO, CI with $\kappa_{\rm H}{}^{\rm N} = 1884  {\rm kg}  {\rm m}^{-2}$	This work
$2.5 \pm 1.0$	local SF	11	$CO(1-0)$ and $\alpha_{CO} = 1$	Jiao et al. (2019)
1.3 ±	local SF	9	CO(1–0) and $\alpha_{CO}$ from S13	Jiao et al. (2019)
$1.6 \pm 0.7$	$z \sim 1.2 \text{ MS}$	11	CO(2–1) and $\alpha_{CO}(Z)$ ( $\langle \alpha_{CO} \rangle = 3$ )	Valentino et al. $(2018)^b$
$2.0 \pm 0.5$	$z \sim 1.2 \text{ MS}$	11	Dust and $\delta_{\text{GDR}}(Z)$ ( $\langle \delta_{\text{GDR}} \rangle = 134$ )	Valentino et al. $(2018)^b$
$3.9 \pm 0.4$	z = 2 - 3 SMGs	14	$CO(4-3)$ , $CO(1-0)$ and $\alpha_{CO} = 1$	Alaghband-Zadeh et al. $(2013)^b$
$8.4 \pm 3.5$	SMGs/QSOs	10	CO(3–2) and $\alpha_{CO} = 0.8$	Walter et al. (2011)
$8.3 \pm 3.0$	local (U)LIRGS	23	CO(1–0) and $\alpha_{CO} = 0.8$	Jiao et al. $(2017, 2019)^b$
$0.9 \pm 0.3$	z = 1 ISM selected	2	CO(2–1), CI and $\alpha_{CO} = 2.6$	Boogaard et al. (2020)
$2.0 \pm 0.4$	z = 1 ISM selected	3	1.2 mm, C I and $\alpha_{850} = 6.7 \times 10^{12}$ from Sco16	Boogaard et al. (2020)
$1.6^{+1.3^{a}}_{-0.7}$	z = 2-4 GRB/QSO absorbers	19	$H_2$ and $C^0$ absorption lines for $Z_{\odot}$	Heintz & Watson (2020)
$7^{+7}_{-35}$	NGC 7469 (CND)	1	AGN, dynamical mass, $L'_{CI}$ and $L'_{CO}$	Izumi et al. (2020)
$1.4-5^{a}$	NGC 6240	1	$\alpha_{\rm CO}$ from CO–SLED, high-density tracers	Cicone et al. (2018)
			And two-phase LVG modelling	Papadopoulos et al. (2014)

*Notes.* <sup>*a*</sup>Indicates estimates of  $X_{CI}$  independent of assumptions for  $\alpha_{CO}$  or  $\kappa_{H}$ .

<sup>b</sup>Indicates that this sample forms part of the literature sample we have used, although we have calibrated  $X_{CI}$  using the submm luminosity and an average normalization of  $\kappa_{H}^{N} = 1884 \text{ kg m}^{-2}$  ( $\delta_{GDR} = 135 \text{ for } \kappa_{850} = 0.071 \text{ m}^2 \text{ kg}^{-1}$ ) for the sample, rather than  $L'_{CO}$  and a fixed  $\alpha_{CO}$ . A breakdown of our results by intensity of star formation can be found in Table 8.

Table 14.	Summary of our em	pirical aco calibrations c	ompared to work in the literature	, $\alpha_{CO}$ is quote	d including a factor of	of 1.36 for He
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$\overline{\alpha_{\rm CO}} M_\odot  ({\rm Kkms^{-1}pc^2})^{-1}$	Sample	Ngal	Notes	Reference
$3.6^{+1.3}_{-1.0}$	$z = 0-5$ , log $L_{\rm IR} > 11$	90	$L_{850}$ , CO, C I with $\kappa_{\rm H}{}^{\rm N} = 1884  {\rm kg}  {\rm m}^{-2}$	This work
$4.2^{+1.8}_{-1.1}$	$z = 0-5, \log L_{\rm IR} > 11$	240	$L_{850}$ , CO with $\kappa_{\rm H}{}^{\rm N} = 1884  {\rm kg}  {\rm m}^{-2}$	This work
$4.8^{+1.3}_{-1.1}$	local MS, log $L_{\rm IR} < 11$	88	$L_{850}$ , CO with $\kappa_{\rm H}{}^{\rm N} = 1884  {\rm kg}  {\rm m}^{-2}$	This work
$3.1^{+3.1a}_{-1.5}$	Local discs	26	$CO(2-1), r_{21} = 0.7, HI, dust$	Sandstrom et al. (2013)
$4.2 (3.5-5.4)^{a}$ $3.4 \pm 2.1^{a}$ $4.1^{+4^{a}}_{-2}$ $2-4^{a}$	MW large scale Planck lensed SMGs NGC 7469 (CND) NGC 6240	24 1 1	$\gamma$ -ray various LVG: multi-J CO, C I and dust AGN, dynamical mass, $L'_{CO}$ LVG: multi-J CO, dense gas tracers	Remy et al. (2017) Harrington et al. (2021) Izumi et al. (2020) Panadopoulos et al. (2014)
$3.8^{+1.0a}_{-0.7}$	z = 0-5	22	CO, CI, Z and absorber based $\alpha_{CI}$	Heintz & Watson (2020)
$4.4^{+2.0^{a}}_{-1.4}$	Local galaxies	24	C II, CO(1–0) and modelling at $Z_{\odot}$	Accurso et al. (2017)
$0.6 \pm 0.2^{a}$ $2-6^{a}$ $3.9 \pm 1.1^{a}$ $1.8 \pm 0.5$ $2.9 \pm 0.5$ $2.9$ $2.4 \pm 0.4$ $1.9 \pm 0.3$ $2.2 \pm 1.0$	(U)LIRGs (U)LIRGs Local discs MW local clouds MW local clouds Taurus Local discs Resolved local discs	28 28 9 6 6 1 7 6	LVG: Single component, multi-J CO LVG: Two-comp, free $dV/dR$ , dense-gas tracers CO(1–0), H I, dust H I, CO(1–0), $\gamma$ -ray H I, CO(1–0), 850- $\mu$ m dust H I, CO(1–0) and extinction/reddening CO(1–0), H I, dust H I, CO(1–0), [C I](1–0), dust, Z, $\kappa_{eff} \sim 1300$ CO(2–1), [C I](1–0), dust, Z, $\kappa_{eff} \sim 1300$	Papadopoulos et al. (2012b) Papadopoulos et al. (2012b) Eales et al. (2012) Remy et al. (2017) Remy et al. (2017) Chen et al. (2015) Cormier et al. (2018) Jiao et al. (2021) Pachwall et al. (2017)
$2.4 \pm 0.4$ $1.9 \pm 0.3$ $3.2 \pm 1.0$	Local discs Resolved local discs z = 4 lensed SMGs	7 6 9	CO(1–0), H I, dust H I, CO(1–0), [C I](1–0), dust, Z, $\kappa_{\text{eff}} \sim 1300$ CO(2–1), [C I](1–0) $X_{\text{CI}} = 3 \times 10^{-5}$	

*Notes*. Errors are  $1\sigma$  standard deviations (or 16–84 percentiles). A breakdown of our results by intensity of star formation can be found in Table 8. <sup>*a*</sup>Indicates estimates which do not rely on assumptions for  $X_{CI}$  or  $\kappa_{H}$ .

LIRGs, with  $L'_{\rm CI}/L'_{\rm CO} = 0.20 \pm 0.04$ . Any study which wishes to test the bi-modality hypothesis must also be representative of the galaxy global properties.

Here, we must stress again that for individual galaxies, joint SLED/SED radiative transfer models of well-sampled SLEDs and dust emission SEDs do recover Galactic-valued  $\alpha_{CO}$  factors even in (U)LIRGs or SMGs (Papadopoulos et al. 2014; Harrington et al. 2021). However, such results cannot be used in a statistical sense, i.e. as typical of the respective galaxy populations for obvious reasons, and our statistical approach remains the sole avenue.

Indeed, the only way that a bi-modal  $\alpha_{CO}$  for MS galaxies and SMGs can be reconciled with  $M_{H2}$  estimates using dust or C1 is to impose the same bi-modality on their conversion parameters ( $\kappa_{H}$ ,  $X_{C1}$ ,  $\alpha_{850}$ ,  $\alpha_{C1}$ ). A reduction of  $\alpha_{CO}$  by a factor 3 times necessitates a

decrease [increase] in  $\kappa_{\rm H}$  [ $X_{\rm CI}$ ] by the same factor. Thus, if  $\alpha_{\rm CO} = 0.8$  is preferred for extreme star-forming galaxies (e.g. Walter et al. 2011), then  $\kappa_{\rm H} = 600$  ( $\delta_{\rm GDR} = 43$  for  $\kappa_{850} = 0.071 \, {\rm m}^2 \, {\rm kg}^{-1}$ ) and  $X_{\rm CI} = 5.3 \times 10^{-5}$  must also be adopted (statistically, for this galaxy population). This discrepancy was previously noted by Bothwell et al. (2017) and Valentino et al. (2018) who found that using  $X_{\rm CI}$ (SMG) =  $3 \times 10^{-5}$  with  $L'_{\rm CI}$  as a tracer resulted in larger gas masses than using  $L'_{\rm CO}$  with the 'ULIRG' value of  $\alpha_{\rm CO} = 0.8$ . Therefore, the popular 'choices' of  $\alpha_{\rm CO} = 0.8$  and  $X_{\rm CI} = 3 \times 10^{-5}$  are incompatible with each other.

Based on our current understanding, there are two plausible physical mechanisms which may cause an increase in  $X_{CI}$  and a decrease in  $\kappa_{H}$  in extreme ISM conditions. The effect of enhanced CR densities on carbon chemistry (Bisbas et al. 2015, 2021; Glover &



**Figure 12.** Results assuming a bi-modal behaviour for  $\alpha_{CO}$ . We run the optimization process with the normalization for SMGs set to  $\alpha_{CO}^{N}(SMG) = 0.8$ , keeping MS galaxies at  $\kappa_{H}^{N}(MS) = 1884 \text{ kg m}^{-2}$  (equivalent to  $\alpha_{CO} = 2.8$ ). This creates the strong bi-modality in  $\alpha_{CO}$  but also induces a similarly strong or even stronger effect in  $\kappa_{H}$  and  $X_{CI}$ , which would be mirrored in  $\alpha_{850}$  and  $\alpha_{CI}$ .

Clark 2016; Gong et al. 2020) favours a higher [C<sup>0</sup>/CO] abundance, however, this mechanism is density dependent and is less effective in dense regions, which typify the ISM of SMGs. Thus, while extreme environments with elevated CRs or X-rays would certainly act to increase  $X_{CI}$  at a fixed density, it does not simply follow that extreme SF activity will produce high  $X_{CI}$  since those same regions (CRDR/XDR) are typically found in regions with increased density.

The higher dense gas fractions common in SMGs may favour higher rates of grain growth, or mantling, both of which would reduce the value of  $\kappa_{\rm H}$  – (i) by decreasing the  $\delta_{\rm GDR}$  and (ii) by increasing the dust emissivity,  $\kappa_{850}$ . Our results imply, however, that any such changes must act in harmony with each other so as to maintain the same observable ratios, so an increase in  $X_{\rm CI}$  must correlate directly with a decrease in  $\kappa_{\rm H}$  and  $\alpha_{\rm CO}$ . This prediction is a clear challenge to models, and full astro-chemical simulations for the extreme physical conditions expected in the ISM of SMGs and ULIRGs will be needed to explore how the three tracers can vary in the exact same way through very different physical mechanisms.

#### 5.4 On the robustness of our choices

#### 5.4.1 Impact of uncorrected $f_{\rm HI} > 1$ galaxies

Statistically, the effect of having uncorrected  $f_{\rm HI} > 1$  galaxies is small, since there are only 15 such galaxies, where  $\langle \alpha_{\rm CO} \rangle$  increases<sup>36</sup> by +0.27 in their luminosity bin compared to when they are removed altogether (by +0.18 compared to when they have been corrected). We are, thus, confident that dust associated with H<sub>I</sub> is not biasing our overall determination of conversion factors and their trends, at least in this sample. For individual galaxies, however, the difference in  $\alpha_{\rm CO}$  can be very large. When using this method for low-redshift galaxies with significant H<sub>I</sub> within the dust-emitting region, corrections are needed.

#### 5.4.2 Impact of using a constant $T_{mw}$

The strong correlation found between  $T_{\rm mw}$  and luminosity (Fig. 1) has not been considered in previous works. In Appendix H2, we show a comparison of results using our empirical  $T_{\rm mw}$  relations to those for constant,  $T_{\rm mw} = 25$  K. Summarizing these findings:

<sup>36</sup>Note that this offset is linear, not logarithmic.

(i) The median offsets between parameters when using constant versus variable  $T_{\rm mw}$  are <0.015 dex. The scatter in a parameter is generally within 0.1 dex (Fig. H2). Thus, the global averages we present in this paper are not affected by a change to constant  $T_{\rm mw} = 25 \,\mathrm{K}$ .

(ii) Allowing  $T_{\rm mw}$  to vary with  $L_{\rm IR}$  is more realistic and leads to a shallow but significant trend with luminosity, such that  $\alpha_{\rm CO}$  decreases with increasing  $L_{\rm IR}$ , while  $\kappa_{\rm H}$ ,  $\alpha_{850}$ , and  $X_{\rm C1}$  increase slightly. Using a constant  $T_{\rm mw}$  of 25 K produces no trends of any conversion factor with  $L_{\rm IR}$  (Fig. H3).

(iii) Using a constant  $T_{\rm mw}$  of 25 K results in gas masses up to 0.1 dex lower at log  $L_{\rm IR} < 11$  and 0.1 dex higher at  $l_{\rm IR} < 12.0$  compared to the variable  $T_{\rm mw}$  used in the main analysis (Fig. H4).

## 6 DISCUSSION

The diverse galaxies in this study show a remarkable consistency in their gas mass tracers, with linear relationships between all three pairs of observables,  $L_{850}$ ,  $L'_{CI}$ , and  $L'_{CO}$ .

We find weak trends in the conversion factors with  $L_{IR}$ ; decreasing  $\alpha_{CO}$ ,  $\alpha_{CI}$  and increasing  $\alpha_{850}$ ,  $\kappa_{H}$ ,  $X_{CI}$ . These trends are very shallow, amounting to a factor <2 times change over 2–3 orders of magnitude in luminosity. The intrinsic variation in  $\kappa_{H}$  and  $X_{CI}$  (the physical quantities encompassing most of the uncertainties in the corresponding conversion factors) is likely very small, and approximating them with a single constant value should be robust. For the sub-samples with a CI tracer (daX, Xa, Xd), we see decreases in all three tracer conversion factors at log  $L_{IR} < 11$ :  $X_{CI}$  (15–25 per cent),  $\alpha_{CO}$  (10–30 per cent), and  $\kappa_{H}$  ( $\delta_{GDR}$ : 20–25 per cent). However, the data indicating a drop in conversion factors originates from the J19 sample (see earlier discussions). More C1 studies of normal star-forming galaxies in the local Universe are urgently required to further explore any such trend, in particular using the global C1 line emission, rather than that of the central few kpc of a galaxy.

The average values of  $X_{CI}$ ,  $\alpha_{CO}$ , and  $\kappa_{H}$  ( $\delta_{GDR}$ ) for galaxies with all three tracers (the daX sample) and log  $L_{IR} > 11$  are our 'reference' values,  $\alpha_{CO}^{R} = 3.7$  (including He),  $X_{CI}^{R} = 1.6 \times 10^{-5}$ ,  $\kappa_{H}^{R} = 1990$  ( $\delta_{GDR}^{R} = 141$ ). These agree within the errors with the mean values determined using only two tracers. These reference values are *not unique* because only the ratios and products of the conversion factors are constrained by the observables,  $L'_{CI}/L'_{CO}$ ,  $L'_{CI}/L_{850}$ , and  $L_{850}/L'_{CO}$ . Once a conversion factor is known or assumed, however, the others can be determined by the self-consistent ratios listed in Table 3. For example, using the ad sub-sample and normalizing to  $\kappa_{\rm H}^{\rm N} = 2800 \,{\rm m}^2 \,{\rm kg}^{-1}$  would produce  $X_{\rm CI} = 1.1 \times 10^{-5}$  and  $\alpha_{\rm CO} = 4.7$ , in reasonable agreement with Accurso et al. (2017) for  $Z = 0.6 \, Z_{\odot}$ , while normalizing to  $\alpha_{\rm CO}^{\rm N} = 0.8$  gives  $X_{\rm CI} = 5.8 \times 10^{-5}$  and  $\delta_{\rm GDR} = 34$ . While the data are consistent with both of these possibilities, or any other combination of the above ratios, we must caution that the low values of  $\alpha_{\rm CO}$  often recovered from CO-only methods (and after modelling only a few low-J CO lines) may be an artefact of well-known gas-dynamics effects, which are expected to have very little impact on the global C1 line emission and none whatsoever on the corresponding dust continuum.

For galaxies at log  $L_{\rm IR} < 11$ , we can only use the ad sample (CO and dust continuum) because of the uncertainties surrounding the C I<sup>cor</sup> galaxies. For the 88 galaxies at log  $L_{\rm IR} < 11$  with CO and dust measurements,  $\alpha_{\rm CO} = 4.8^{+1.4}_{-1.1}$  (including He), with  $\langle \kappa_{\rm H} \rangle = 1718 \,\mathrm{kg \, m^{-2}} (\delta_{\rm GDR} = 122)$  but note that this is still a sample of massive and metal-rich galaxies, just at lower log  $L_{\rm IR} \sim 9-11$ . This study is not applicable to low mass metal-poor galaxies.

## 7 CONCLUSIONS

We have cross-calibrated the three mainstays of molecular gas measurements in extra-galactic astronomy: <sup>12</sup>CO(1–0), [C I](<sup>3</sup> $P_1$ –<sup>3</sup> $P_0$ ) and submm continuum emission from dust. This analysis uses galaxy samples spanning 0 < z < 5 and more than four orders of magnitude in  $L_{\rm IR}$ . All the galaxies are metal rich and/or massive, to remove the need for large corrections for metallicity effects.

(i) We present a new method of optimizing gas mass estimation when multiple tracers are observed, making use of the intrinsic scatter in all three pairs of gas tracers. We demonstrate its effectiveness compared to the simpler method used previously in the literature, and give examples and prescriptions for its use.

(ii) In a purely empirical analysis, we show that  $L'_{CI}$  is the molecular gas tracer with the least intrinsic scatter, particularly at log  $L_{IR} > 11$ . In such galaxies,  $L'_{CI}$  should be the preferred tracer, all other considerations being equal.

(iii) Using our optimized method, we determine the mean empirical conversion factors for  $M_{mol}$  (including He). For log  $L_{IR} > 11$  these are:  $\alpha_{CI}^{R} = 17.0 \pm 0.3$ ,  $\alpha_{850}^{R} = (6.9 \pm 0.1) \times 10^{12}$ ,  $\alpha_{CO}^{R} = 4.0 \pm$ 0.1, with a scatter of 0.11–0.15 dex. These values are for an overall normalization set to the average dust properties of local galaxies and diffuse dust in the Milky Way ( $\kappa_{H} = \delta_{GDR}/\kappa_{850} = 1884 \text{ kg m}^{-2}$ ). A change in this choice of normalization will affect  $\alpha_{CO}$  and  $\alpha_{CI}$  in a proportional manner and  $\alpha_{850}$  in an inversely proportional way. Our reference conversion values can be applied to any metal-rich galaxy with  $Z > 0.5 Z_{\odot}$  in the range 0 < z < 6.

(iv) Using the same method, we determine the principal mean physical parameters on which these conversion values depend. For galaxies at log  $L_{\rm IR} > 11$ :  $X_{\rm C1}^{\rm R} = 1.6 \times 10^{-5}$ ,  $\kappa_{\rm H}^{\rm R} = 1990 (\delta_{\rm GDR}^{\rm R} = 141)$ .

(v) The relationships between the observables,  $L_{850}$ ,  $L'_{CO}$ , and  $L'_{CI}$  are consistent with being linear and the ratios of these observables do not show a strong dependence on IR luminosity, dust temperature, redshift, or the intensity of star formation.

(vi) The ratio of  $L'_{CI}/L'_{CO}$  is marginally (3 $\sigma$ ) different for MS galaxies and SMGs, with the latter having higher  $L'_{CI}/L'_{CO}$ , broadly consistent with expectations from astro-chemical cloud models that include enhanced CRs.

(vii) We find  $Q_{10} = 0.48$  to be a reasonable choice for the excitation function (required to convert  $L'_{CI}$  to  $M_{CI}$ ), based on recent

analysis showing that  $Q_{10}$  has a superthermal behaviour in non-LTE conditions (Papadopoulos et al. 2022). For a range of plausible galaxy ISM density and gas temperatures, the 99th percentile confidence interval on this value is  $\pm 16$  per cent.

(viii) We present empirical relations for the mass-weighted dust temperature,  $T_{\rm mw}$ , to allow observers to better estimate their dust calibration factors. We find a significant trend, where  $T_{\rm mw}$  increases with  $L_{\rm IR}$ . The median  $T_{\rm mw}$  for SMGs at  $z \sim 2.5$  is  $T_{\rm mw}^{\rm SMG} = 30.1 \pm 0.7$  K, while for MS galaxies  $T_{\rm mw}^{\rm MS} = 23.0 \pm 0.4$  K.

(ix) We find a weak trend for  $\kappa_{\rm H}$  and  $X_{\rm CI}$  to increase with  $L_{\rm IR}$ , and a similar trend for  $\alpha_{\rm CO}$  to decrease. The empirical conversion factors ( $\alpha_{\rm CO}$ ,  $\alpha_{\rm CI}$  and  $\alpha_{\rm 850}$ ) also show a shallow but significant correlation with their tracer luminosities. These trends are not apparent if a constant  $T_{\rm mw}$  is adopted. They are therefore driven by the change in  $T_{\rm mw}$  with luminosity.

(x) Using an Occam's Razor assumption that metal-rich galaxies have similar dust emissivity per unit gas mass, we find no evidence for the factor 3–4  $\alpha_{\rm CO}$  bi-modality between SMGs and MS galaxies often adopted in the literature. The shallow trends we do find reflect the common assumption that extreme SF systems have lower  $\alpha_{\rm CO}$  and higher  $X_{\rm CI}$ , albeit at a far more subtle level, with only a ~15 per cent difference in the sample mean  $\alpha_{\rm 850}$  (higher),  $\alpha_{\rm CI}$  (lower) and  $\alpha_{\rm CO}$  (lower) for extreme star-forming galaxies versus 'normal' MS star-formers. o overall

(xi) With the Occam's Razor assumption, we also find no evidence to support the extremely high global estimates of  $X_{CI}$  (~6 × 10<sup>-5</sup>) reported in some literature for ULIRGs/SMGs – the high reported values are a consequence of assuming a low  $\alpha_{CO} \sim 1$ . High  $X_{CI}$ values may be expected, and indeed have been measured in small (<500 pc) regions such as M82 (nuclear starbursts) and XDR regions around AGN, but the extent to which a global estimate would be enhanced depends on the dominance of that extreme environment in the galaxy's H<sub>2</sub> reservoir.

(xii) One can, however, still postulate a different prior for the normalization assumption and impose the popular bimodality in  $\alpha_{CO}$ . The constancy of the measured tracer luminosity ratios then forces the conversion factors for the other two tracers (dust and C1) to become bi-modal in the same way.

We conclude by noting that lacking a direct  $M_{\rm H2}$  measurement method (i.e. via the H<sub>2</sub> lines themselves), one must assume a normalization for one of the sample mean conversion factors in statistical studies like ours. In this study, we choose to benchmark to the dust emission, with  $\kappa_{\rm H}^{\rm N} = 1884 \, \rm kg \, m^{-2}$ . Other normalization choices can, of course, be made, but currently dust emission is the simplest and best understood tracer, and has the advantage of being totally insensitive to the gas-dynamic effects that affect the  $\alpha_{CO}$ conversion factor (e.g. unbound molecular gas components in the winds that exist in actively star-forming galaxies; winds which can be CO-bright while carrying little mass). The [C I](1-0) line emission will also be largely unaffected by these gas-dynamics effects, and as such the corresponding conversion factor,  $\alpha_{CI}$ , shows promise as a good benchmark, borne out by the empirical finding that it has the least intrinsic scatter of the three tracers. With more extensive observational and theoretical studies of CI line emission (particularly in galaxies of lower IR luminosity), the limits of its usefulness as a gas tracer can be determined.

#### ACKNOWLEDGEMENTS

The authors thank the referee for their careful reading and insightful comments on the original version of the paper. LD thanks P. Clark, S. Glover, Q. Jiao, and T. Bisbas for helpful discussions. LD, SJM,

Consolidator grant, Cosmicdust. This paper makes use of the following software available publicly from github: corner.py, emcee.py (Foreman-Mackey et al. 2013b; Foreman-Mackey 2016).

## DATA AVAILABILITY

Data tables based on the samples used in this paper are available via anonymous ftp to cdsarc.u-strasbg.fr (130.79.128.5), alternatively via http://cdsarc.u-strasbg.fr/viz-bin/qcat?J/MNRAS/. The data sets were derived from sources in the public domain, which are listed in Table 1.

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## APPENDIX A: NOTES ON THE LITERATURE FLUXES

In order to produce a homogeneous and up-to-date set of fluxes, we have applied the following corrections.

(i) Since Sco16 was published, the 500- $\mu$ m flux densities used

for their local sample (Dale et al. 2012) were updated following the

latest Herschel calibration. To estimate  $T_{\rm mw}$ ,  $T_{\rm d}$ , and  $L_{850}$ , we fitted

the photometry presented by Chu et al. (2017) and Clark et al. (2018)

MNRAS 517, 962-999 (2022)

#### A1 Corrections to previously published work

using the method described in Dunne & Eales (2001).

(ii) The 850- $\mu$ m photometry for local galaxies in the SLUGS sample (Dunne et al. 2000) is contaminated by the CO(3–2) line. We have corrected for this using the results of Seaquist et al. (2004), where for galaxies with D < 148 Mpc, we reduce the 850- $\mu$ m flux density by 25 per cent.

(iii) It appears that the CO(2–1) data from Aravena et al. (2016), as reproduced in Bothwell et al. (2017), has been incorrectly converted to  $L'_{10}$  ( $L'_{21}$  appears to have been multiplied by 0.9 instead of being divided by it). We have corrected this error and applied our chosen value of  $r_{21} = 0.8$  for the conversion.

#### A2 Homogenization of distances

The most local galaxies (D < 30 Mpc) often have a variety of distances used in the literature. As we have often taken  $L_{\text{IR}}$ ,  $L'_{\text{CI}}$ ,  $L'_{\text{CO}}$ , and  $L_{850}$  from different papers, we have had to homogenize the literature luminosities to correspond to a common distance. The distance chosen is that listed in Dale et al. (2017) and presented in Table 1.

#### A3 Updating local CO data

The Sco16 local galaxy sample used CO(1-0) fluxes from the FCRAO single-dish survey of Young et al. (1995), which has significant and uncertain extrapolations to total fluxes for extended galaxies. We have updated the CO data for these very local galaxies to use CO(1-0) maps from the COMING survey (Sorai et al. 2019) where possible as well as from other mapping data sets from the literature (Gao & Solomon 2004; Kuno et al. 2007; Young et al. 2008; Galametz et al. 2011; Koda et al. 2011; Schruba et al. 2012; Ueda et al. 2014).

*New CO measurement for ID141* We use an unpublished CO(1–0) flux for ID141, which was observed with the Jansky Very Large Array and has  $S_{10} = 0.61 \pm 0.09$  Jy km s<sup>-1</sup>.

## **APPENDIX B: REQUIRED CORRECTIONS**

#### B1 H I-dominated galaxies at lower L<sub>IR</sub>

There is a potential source of bias when deriving calibration factors involving  $L_{850}$  for galaxies with large ratios of  $f_{\rm HI} = {\rm H~I/H_2}$ , as the dust may be tracing HI as well as H<sub>2</sub>. If we apply our method from Section 3 to such HI-dominated galaxies, we will infer the presence of more H<sub>2</sub> due to the dust which resides only in the HI phase. Because, we calibrate in pairs of tracers, this leads to an overestimate of  $\alpha_{\rm CO}$  or  $\alpha_{\rm CI}$  as well as a bias in the dust-based calibration factor.

To investigate this, we estimated  $f_{\rm HI}$  in the same regions as the submm flux densities for the local galaxies we could find in the literature (Dunne et al. 2000; Spekkens, Irwin & Saikia 2004; Wong et al. 2013; Groves et al. 2015; Thuan et al. 2016; Dale et al. 2017; Koribalski et al. 2018; Jiao et al. 2021). As  $f_{\rm HI}$  correlates inversely with  $M_*$ , metallicity, and  $L_{\rm IR}$  (e.g. Bothwell et al. 2014; Saintonge et al. 2016), this issue affects more of the low  $L_{\rm IR}$  galaxies (mostly in the ad sample). For any galaxies with  $f_{\rm HI} > 1$  within the optical disc, we make a correction to  $L_{850}$ , removing that portion of the dust emission which is likely associated with the excess H I. This correction is designed to produce the same  $L_{850}/\rm{H}_2$  ratio as a galaxy with  $f_{\rm HI} = 1$ .

$$L_{850}^{\rm cor} = L_{850} \left(\frac{2}{f_{\rm HI} + 1}\right). \tag{B1}$$

Galaxies with  $f_{\rm H\,I} > 1$  are shown with this correction applied as cyan diamonds in the figures. The higher luminosity (U)LIRGs and SMGs are dominated by molecular gas (e.g. Yao et al. 2003), so we do not need to correct these.

#### B2 Discussion of local C1 data

For the *Herschel* FTS measurements of local (U)LIRGs (Lu et al. 2017), we only include local galaxies with D > 27 Mpc to avoid issues with mis-matched beams. We also rejected galaxies where there was a large discrepancy between the measurement of Lu et al. (2017) and that of Kamenetzky et al. (2016; using the same data).

The set of local galaxies which were mapped by the *Herschel* FTS and presented by J19 are shown in the figures, but not included in the averages for the following reasons:

(i) The CI and CO measurements are made in matched apertures, however, the area mapped in CI is sometimes much smaller than that used for the 500–850- $\mu$ m flux densities reported in the literature. Any analysis which involves both  $L'_{CI}$  and  $L_{850}$  requires a correction to  $L'_{CI}$  to address the mis-match in apertures. We attempted to do this by taking the global CO luminosities (which are equivalent global fluxes to the submm continuum measurements) and assume that the deficit between the global  $L'_{CO}$  and that measured in the same aperture as the CI by J19 is the same as the deficit in  $L'_{CI}$ 

$$L_{\rm CI}^{\prime \ \rm cor} = L_{\rm CI}^{\prime \ \rm J19} \frac{L_{\rm CO}^{\prime \ \rm global}}{L_{\rm CO}^{\prime \ \rm J19}}.$$
 (B2)

These corrections (JC) range from JC = 0.00–0.74 dex, and the pink diamonds in the figures indicate those galaxies that have JC > 0.07 dex. Even after applying the corrections, the J19 galaxies have different average properties in the  $L_{850}/L'_{C1}$  ratio (see Fig. 2). We therefore, do not have confidence in our comparison of  $L'_{C1}$  to  $L_{850}$  for these galaxies and so exclude them from the statistics.

(ii) Although the CO and C I luminosities from J19 are measured in the same apertures, there is a trend for these resolved galaxies to have lower  $L'_{CI}$  for a given  $L'_{CO}$  compared to galaxies which have more global flux measurements. There could be a sampling bias because C I is only detected over the inner kpc or so of the larger galaxies. The CO luminosity per mass of gas ( $\alpha_{CO}$ ) has been found to be lower in the central regions of many galaxies (Sandstrom et al. 2013), which would produce a decrease in  $L'_{CI}/L'_{CO}$ . Since we wish to compare the same averaged global fluxes across all galaxies, we remove these 'centrally-biased' galaxies from our statistical analysis, but we show them in the figures for completeness.

(iii) Finally, a more recent paper by Jiao et al. (2021) did produce matched dust and C I measurements for a subset of the J19 galaxies. The results are shown in Fig. 3(d) where it can be seen that the J21 galaxies are still deficient in C I compared to the higher luminosity galaxies. This cannot be due to a mis-matched aperture but the same sampling bias is present toward the inner regions of the resolved galaxies. An offset to lower  $L'_{CI}$  per  $L_{850}$  implies either depressed  $L'_{CI}$  (lower  $X_{CI}$ ) or increased  $L_{850}$  per unit gas mass (lower  $\delta_{GDR}$ , or higher dust emissivity).

(iv) Unfortunately, this is the only published set of CI fluxes for galaxies with log  $L'_{\rm CI} < 8$  and the only set of fluxes published for the mapping mode of the *Herschel* FTS. There is no description in the literature of how the processing for this mode should be made, and there are differences in the results of J19 and Crocker et al. (2019), who analyse some of the same mapping data. Despite our best attempts to contact the relevant team, we have not been given the details of their flux measurements. We can only note that the

C I fluxes from *Herschel* FTS mapping are not necessarily repeatable when analysed by different teams and so elect to exclude the resolved J19 galaxies from the statistical analysis.

Excluded galaxies are denoted as 'C I<sup>cor'</sup> and they are shown as pink diamonds on the relevant figures.

## APPENDIX C: DUST MASS OPACITY AND THE RELATIONSHIP OF DUST TO GAS

The dust mass opacity coefficient,  $\kappa_d(\lambda)$ , is proportional to the emissivity per unit mass of dust. It is related to the calibration parameter we use in our analysis,  $\kappa_H = \delta_{GDR}/\kappa_d$ , where  $\kappa_H$  refers to the dust emission per H mass, thus encompassing the two unknowns of dust optical properties and gas-to-dust ratio ( $\delta_{GDR}$ ).

The dust optical properties are not easily measured, and can vary enormously from laboratory-based studies to theoretical dust models and from those inferred by observations (for a review see e.g Dunne et al. 2003; Clark et al. 2019b).

Commonly adopted extragalactic estimates range from  $\kappa_{850} = 0.03-0.08 \text{ m}^2 \text{ kg}^{-1}$  (Dunne et al. 2000; Li & Draine 2001; James et al. 2002; Draine 2003; Planck Collaboration XIX 2011; Eales et al. 2012; Clark et al. 2016; Bianchi et al. 2019), though higher values (by factors of several) are inferred for the very densest and coldest environments where grains can grow icy mantles and coagulate (Köhler et al. 2015; Remy et al. 2017; Ysard et al. 2018). These changes in opacity have also been correlated with a loss of PAH and stochastically heated small grains (Flagey et al. 2009;

Ysard et al. 2013). Remy et al. (2017) suggest that regions of the ISM with dust opacities a factor ~2 higher than the diffuse ISM (and with cold dust,  $T_d \sim 16-18$  K), would be those where grains are accreting carbonaceous mantles, as in the THEMIS dust model (Jones et al. 2017; Jones 2018). This carbon mantle-accreting regime is largely assumed to be the dark neutral medium (close to the atomic-molecular transition, where there is low CO emission and high H<sub>I</sub> opacity). Deeper within clouds, where the temperature drops to  $T_d < 16$  K, the dust begins to aggregate and accrete ice mantles, which increases the opacity further. These very dense, cold environments do not, however, contain the bulk of the ISM mass and certainly do not emit a dominant fraction of  $L_{850}$  in a galaxy (Draine et al. 2007; Bianchi et al. 2019). The increase in dust emissivity ( $\kappa_{850}$ ) from atomic to moderately dense molecular material is in the range 1.2–2.0 (Remy et al. 2017).

In fact, it is  $\kappa_{\rm H}$  – the parameter relating the dust emissivity to the gas mass – that can be measured in astrophysical situations, since we have no absolute knowledge of  $\delta_{\rm GDR}$ . Table C1 lists a comprehensive set of observational and theoretical values for  $\kappa_{\rm H}$  from the literature. Estimates of  $\kappa_{\rm H}$  in the Milky Way are made across a number of sightlines, from H I-only (diffuse) to H<sub>2</sub>-dominated clouds (dense), where CO emission is used with assumptions about  $\alpha_{\rm CO}$  in order to determine  $N_{\rm H}$ . Independent confirmation is provided by studies (e.g. Remy et al. 2017) using  $\gamma$ -ray observations to determine the gas column; the resulting values of  $\kappa_{\rm H}$  are in good agreement (see Table C1), with  $\kappa_{\rm H}$  being higher along diffuse sightlines (1800–2400), dropping to 700–1500 in denser molecular or dark neutral media.

**Table C1.** Summary of our physical dust calibrations ( $\kappa_{850}$ ,  $\delta_{GDR}$ ) compared to other work in the literature, where  $\delta_{GDR}$  and  $\kappa_{H}$  refer to the mass of hydrogen in all forms, excluding He.

$\frac{\kappa_{\rm H} = \delta_{\rm GDR} / \kappa_{850}}{\rm kg  m^{-2}}$	Sample	Notes	Reference
1884 (1500–2200)	Ex-gal	Average of extragalactic estimates	This work
		Milky Way diffuse and atomic regions	
$2352\pm198$	Diffuse	$850\mu\text{m}$ , H I very diffuse sightlines	Planck Collaboration XVII (2014)
$1988\pm710$	All sky	850 $\mu$ m, H I, CO(1–0) with $\alpha_{CO} = 3.2$	Planck Collaboration XI (2014)
$1380\pm251$	Taurus HI	H I with 25-per cent opacity correction, Planck, scaled $\beta = 1.8$	Planck Collaboration XIX (2011)
1518		250 $\mu$ m scaled to 850 mic with $\beta = 1.8$ , H I	Boulanger et al. (1996)
		Milky Way molecular/higher density regions	
1392	$\log(N_{\rm H}) > 20$	850 $\mu$ m, H I, CO(1–0) with $\alpha_{CO} = 3.2$	Planck Collaboration XI (2014)
1392	$\log(N_{\rm H}) \sim 21$	850 $\mu$ m, H I, CO(1–0) with $\alpha_{\rm CO}^{\gamma}$	Remy et al. (2017)
1012-1044	DNM	Dark neutral medium, 850 $\mu$ m, $\gamma$ -rays	Remy et al. (2017), Remy et al. (2018)
$700 \pm 200^a$	Local clouds (H <sub>2</sub> )	850 $\mu$ m, CO(1–0), $\alpha$ <sub>CO</sub> from $\gamma$	Remy et al. (2017)
$1210 \pm 184^{a}$	Local clouds (H I)	850 μm, H I	Remy et al. (2017)
$654\pm85$	Taurus H <sub>2</sub>	NIR extinction, Planck, scaled $\beta = 1.8$	Planck Collaboration XIX (2011)
		Local galaxies	
$1663\pm333$	9	CO(1–0), H I, 500 $\mu$ m dust scaled to 850 $\mu$ m with $\beta = 1.8$	Eales et al. (2012)
2296 (163/0.071)	101 Sab-Sbc	CO(1–0) with $\alpha_{CO} = 3.2$ , H I, dust SED fits	Casasola et al. (2020)
1692-2169	130 Sa-Sc	CO(1–0), H I, dust MBB, $\alpha_{CO}(Z)$	Bianchi et al. (2019)
2096	26	CO(2–1), H I, dust DL07 fits	Sandstrom et al. (2013)
2402 (92/0.0383)	189	CO(1–0), H I, dust DL07 fits $\alpha_{CO} = 3.2$	Orellana et al. (2017)
1500-2200	M74, M83	Z, H I, CO(2–1), 500 $\mu$ m with James et al. (2002) method	Clark et al. (2019b)
		Physical dust models commonly used in the literature.	
3232 (109/0.034)	Theoretical	Physical dust model producing too much $A_v/N_{\rm H}$	Draine (2003); DL07
			Planck Collaboration XXIX (2016)
1972	Theoretical	Up-dated DL07 dust model	Draine & Hensley (2021)
1901 (135/0.071)	Theoretical	Physical dust model THEMIS	Jones et al. (2017), Jones (2018)

*Notes.* The first column is  $\kappa_{\rm H}$ , the ratio of the gas-to-dust ratio ( $\delta_{\rm GDR}$ ), and the dust mass opacity coefficient. Where there is an explicit assumption for  $\delta_{\rm GDR}$  or  $\kappa_{850}$  in a reference, we include it in parentheses. <sup>*a*</sup> The clouds in these rows are the same; Remy et al. have calculated the dust opacity for each gas phase separately.  $\alpha_{\rm CO}(Z)$  from Amorín et al. (2016).

In extragalactic studies, a similar method is used, although with larger uncertainties as it is less straightforward to decompose the atomic and molecular components along the line of sight. These studies find a range of  $\kappa_{\rm H} = 1500{-}2200 \,\rm kg \,m^{-2}$ , closer to the diffuse ISM measurements in the Milky Way.

For a given dust model, we can also calculate the theoretical  $\kappa_{\rm H}$  given the assumed dust optical properties, chemical abundances, and depletions. The theoretical values are also listed in Table C1 where the current consensus is for  $\kappa_{\rm H} \sim 1900-2000 \,\rm kg \,m^{-2}$ . The popular Draine (2003) model has a significantly higher  $\kappa_{\rm H} = 3200 \,\rm kg \,m^{-2}$  (lower  $\kappa_{850} = 0.034 \,\rm m^2 \,\rm kg^{-1}$  for  $\delta_{\rm GDR} = 109$ ) than all of the empirical measurements. This was noted by Draine et al. (2014) and Planck Collaboration XXIX (2016) and has been updated in the more recent version of this model by Hensley & Draine (2021). We encourage readers to use the updated version in order to produce dust-based measurements which are consistent with what we know about dust from observations.

## APPENDIX D: DERIVING GAS MASS FROM OBSERVATIONS OF $[C I]({}^{3}P_{1} - {}^{3}P_{0})$

The excitation term,  $Q_{ul}$ , which describes the fraction of C atoms in each excited state, is a function of  $(n, T_k)$  in non-LTE conditions, and is derived analytically in the Appendix to Papadopoulos et al. (2004). A recent study of the [CI](2-1)/(1-0) line ratio found that [CI](2-1)is strongly sub-thermally excited, and [CI](1-0) presents interesting superthermal behaviour in the range of density and temperature expected for galaxies. We illustrate the dependence of  $Q_{ul}$  on  $(n, T_k)$  in Fig. D1. As discussed by Papadopoulos et al. (2022), the value of  $Q_{10}$ for lower densities  $(n = 300-3000 \text{ cm}^{-3})$  can exceed the LTE value at  $T_k > 20$  K, but Fig. D1 shows that for a reasonable range of n and  $T_{\rm k}$  (300 < n < 10 000 cm<sup>-3</sup>, 25 <  $T_{\rm k}$  < 80 K)  $Q_{10}$  does not go outside the range 0.35–0.53. In fact, for a uniform probability of  $(2.4 < \log n)$ < 4.0) and (25 <  $T_{\rm k}$  < 80 K) the 99-per cent range for  $Q_{10}$  is 0.40-0.54, median = 0.48. The relative uncertainty on the calibration of C I mass from the lack of knowledge of  $(n,T_k)$  is, thus,  $<\pm 16$  per cent. We will therefore use the median value of  $Q_{10} = 0.48$  throughout, because even though we may be able to use the measured  $T_d$  to infer the galaxies with higher or lower  $T_k$  (assuming  $T_k = \alpha^{\text{TD}} T_d$  – see Papadopoulos et al. 2022), the lack of knowledge of the density and the superthermal behaviour in the J = 1 state means that there is no direct correlation between  $Q_{10}$  and  $T_k$ . Using sensible average

parameters for MS galaxies [and SMGs], so n = 500 [5000] cm<sup>-3</sup> and  $T_k = 40$  [80] K, we find only a small (~ 10 per cent) difference in the  $Q_{10}$  values expected.

The LTE expressions for  $Q_{10}$  and  $T_x$  should not be used (Papadopoulos et al. 2022) as  $Q_{10}^{LTE}$  is actually *lower* than the non-LTE  $Q_{10}$  for densities higher than a few hundred cm<sup>-3</sup>, and, thus, its use would lead to a systematic bias – e.g. for  $T_k = 60$  K and n = 1000cm<sup>-3</sup>, the LTE value for  $Q_{10}$  is 18 per cent lower than the appropriate non-LTE value. This would lead to an 18-per cent overestimate of the H<sub>2</sub> mass using C I.

For the [C1](2–1) line, things are not so promising [Fig. D1(righthand panel)]. The range of possible values of  $Q_{21}$  are large, ranging from 0.07–0.37 for the 99-per cent range. The median is  $Q_{21} = 0.22$ , giving an uncertainty range of ±68 per cent for reasonable values of  $(n,T_k)$ . Because of the sub-thermal behaviour, the [C1](2–1) line is a sensitive indicator of density (Papadopoulos et al. 2022) and galaxies with strong [C1](2–1) emission will have a larger fraction of their H<sub>2</sub> in a dense state.

## APPENDIX E: FROM PAIRWISE VARIANCES TO INDIVIDUAL VARIANCES

We have measurements of different tracers of gas mass for several galaxies, but no direct measurements of  $M_{\rm H2}$  itself. Hence, it is not possible to measure directly how well each tracer follows the gas mass. However, we do have measurements of the different tracers for each galaxy, so we can estimate the scatter in the difference between the tracers. Under some assumptions this allows us to infer the scatter between each tracer and the gas mass.

To simplify the notation, we write the log of observed quantities and corresponding standard deviation of errors as

$$\begin{aligned} x_1 &= \log(L_{850}), & \sigma_1 &= \log(1 + \sigma_{850}/L_{850}), \\ x_2 &= \log(L'_{CO}), & \sigma_2 &= \log(1 + \sigma_{CO}/L'_{CO}), \\ x_3 &= \log(L'_{C1}), & \sigma_3 &= \log(1 + \sigma_{C1}/L'_{C1}). \end{aligned}$$
(E1)

If true value of the log of the gas mass is  $\hat{m} = \log(M_{\rm H2})$ , and the true values of the observed quantities are  $\hat{x}_i$ , where i = 1...3, then we can write

$$\hat{m} = \hat{x}_i + \hat{a}_i,\tag{E2}$$



Figure D1.  $Q_{10}$  (left-hand panel) and  $Q_{21}$  (right-hand panel) as function of gas temperature for a range of densities, where lines are labelled with the values of log *n*. The LTE value is shown as the black dash-dot line.

$$\hat{a}_{1} = -\log(\hat{\alpha}_{850}),$$
  
 $\hat{a}_{2} = \log(\hat{\alpha}_{CO}),$   
 $\hat{a}_{3} = \log(\hat{\alpha}_{CI}).$  (E3)

Note that the true values  $\hat{a}_i$  may be different for each galaxy, depending on the individual physical conditions within the galaxies.

If we choose a particular set calibration factors for all galaxies, say  $\tilde{a}_i$ , this provides three estimates of the gas mass for each galaxy

$$m_i = x_i + \tilde{a}_i. \tag{E4}$$

The error in each mass estimate is

$$m_i - \hat{m} = x_i - \hat{x}_i + \tilde{a}_i - \hat{a}_i$$
  
=  $\delta x_i + \delta a_i$ , (E5)

where  $\delta a_i$  is the difference between the true factor for this galaxy and the value we have chosen, and  $\delta x_i$  are the measurement errors of the observations. If we assume that the errors on  $x_i$  are not correlated with the errors on  $a_i$ , then the variance of the mass errors is given by

$$\operatorname{var}(m_i - \hat{m}) = \sigma_i^2 + s_i^2, \tag{E6}$$

where  $s_i^2$  is the variance of the true calibration factors.

The value of  $s_i^2$  gives a direct measure of how accurate the particular tracer is when using a universal calibration factor for all galaxies. Without knowing the true gas mass, we do not have a direct measure of this value, but we can obtain an estimate by considering the differences between the mass measurements

$$m_i - m_j = x_i - x_j + \tilde{a}_i - \tilde{a}_j$$
  
=  $\delta x_i - \delta x_i + \delta a_i - \delta a_j.$  (E7)

If we ignore all co-variance terms, the variance of the differences is given by

$$v_{ij} = \operatorname{var}(m_i - m_j) = \sigma_i^2 + \sigma_j^2 + s_i^2 + s_j^2.$$
 (E8)

It is straightforward to re-arrange these equations to find the intrinsic variance of the calibration factors as

$$s_0^2 = (v_{01} - v_{12} + v_{20})/2 - \sigma_0^2,$$
(E9)

with similar equations for  $s_1^2$ , and  $s_2^2$ . So long as we have good estimates of the measurement errors,  $\sigma_i$ , for the observed quantities, we can estimate the scatter in calibration constants for each tracer. Using our data set, we have measured the variance for each pair of factors in equation (E8). Assuming that the co-variance between the calibration factors is zero, we use the three pair variances to estimate the intrinsic variance of the three individual calibration factors. The resulting standard deviations are  $s_{\kappa} = 0.1294$ ,  $s_{\alpha} = 0.1436$ , and  $s_X = 0.1125$ , using all galaxies except the C I<sup>cor</sup>.<sup>37</sup> Values are listed in Table 3.

This analysis shows that  $X_{CI}$  has the smallest scatter between galaxies, especially when considering log  $L_{IR} > 11$  galaxies, which is a new result, independent of any assumptions.

## APPENDIX F: A BAYESIAN APPROACH TO COMBINING GAS MASS ESTIMATES

Our method of combining the three gas mass tracers is based on the idea that the conversion factors for any particular galaxy come from

<sup>37</sup>When restricting the analysis to log  $L_{\rm IR} > 11$  galaxies, C I produces notably less scatter than both CO and dust continuum, with  $s_{\kappa} = 0.1339$ ,  $s_{\alpha} = 0.1646$  and  $s_{\rm X} = 0.082$ .

parent distributions with variances as derived in Appendix E. This means that we should allow for the expected scatter in conversion factors as well as the observational error when combining estimates from the different tracers. Using a Bayesian approach to the problem, we show the most likely mass estimate is simply the inverse variance weighted mean of the tracers, where the weights include both measurement error and the variance in conversion factors.

We continue to use the notation as in Appendix E, where the observed quantities are  $x_i$  and errors  $\sigma_i$ . Assuming the measurement errors are Gaussian the probability of measuring the observed value of  $x_i$  is

$$P(x_i|\hat{x}_i, \sigma_i) = \mathcal{N}(x_i|\hat{x}_i, \sigma_i^2), \tag{F1}$$

where  $\mathcal{N}$  represents the normal distribution centred on  $\hat{x}_i$  and with variance  $\sigma_i^2$ . Now, for each observation we can use Bayes theorem to estimate the posterior probability that the gas mass is *m* 

$$P(m, \hat{a}_i | x_i) = P(x_i | m, \hat{a}_i) P(\hat{a}_i) P(m) / P(x_i),$$
(F2)

where we have assumed *m* and  $\hat{a}_i$  are independent. For the prior on  $\hat{a}_i$ , we assume a normal distribution with mean  $\bar{a}_i$  and variance  $s_i^2$ , as discussed in Appendix E. We assume a flat prior on *m*, implying that P(m) is constant. Since  $P(x_i)$  is also constant, the position of the maximum posterior probability does not depend on the actual value of  $P(m)/P(x_i)$ , and for convenience we set this to 1. Therefore

$$P(m, \hat{a}_i | x_i) \propto P(x_i | m, \hat{a}_i) P(\hat{a}_i)$$
  
=  $\mathcal{N}(x_i | m - \hat{a}_i, \sigma_i^2) \mathcal{N}(\hat{a}_i | \bar{a}_i, s_i^2)$   
=  $\mathcal{N}(\hat{a}_i | m - x_i, \sigma_i^2) \mathcal{N}(\hat{a}_i | \bar{a}_i, s_i^2).$  (F3)

Here, we have used equation (E2) to go from  $m - \hat{a}_i$  to  $m - x_i$ . Since we are interested primarily in the value of the gas mass, and not explicitly in the values of the calibration factors, we can marginalize over the values of  $\hat{a}_i$ . Ignoring the uncertainties on the variances,  $\sigma_i^2$ and  $s_i^2$ , leads to

$$P(m|x_i) = \mathcal{N}(m|x_i + \bar{a}_i, \sigma_i^2 + s_i^2).$$
(F4)

Including all three observations for the galaxy this becomes

$$P(m|\{x_i\}) = \prod_{i=1}^{3} \mathcal{N}(m|x_i + \bar{a}_i, \sigma_i^2 + s_i^2)$$
  

$$\propto \exp\left(-\sum_{i=1}^{3} \frac{(m - x_i - \bar{a}_i)^2}{2(\sigma_i^2 + s_i^2)}\right).$$
(F5)

So maximizing the posterior probability with respect to *m* is equivalent to minimizing  $\chi^2$ , where

$$\chi^2 = \sum_{i=1}^3 \frac{(m - x_i - \bar{a}_i)^2}{2(\sigma_i^2 + s_i^2)}.$$
 (F6)

The minimum with respect to *m* is given by

$$m^{\text{opt}} = \left(\sum_{i=1}^{3} \frac{x_i + \bar{a}_i}{\sigma_i^2 + s_i^2}\right) / \left(\sum_{i=1}^{3} \frac{1}{\sigma_i^2 + s_i^2}\right)$$
$$= \left(\sum_{i=1}^{3} (x_i + \bar{a}_i)w_i\right) / \left(\sum_{i=1}^{3} w_i\right), \quad (F7)$$

where  $w_i = 1/(\sigma_i^2 + s_i^2)$ . So the optimal mass estimate is simply the inverse variance-weighted mean of the three estimates, where each uses the mean conversion factor, and where the variance for each measure is the sum of the measurement error and the expected variance of the conversion factor. The uncertainty on  $m^{\text{opt}}$  is the uncertainty on the weighted mean

$$\sigma_{m^{\text{opt}}} = 1 \bigg/ \left( \sum_{i=1}^{3} w_i \right). \tag{F8}$$

The corresponding estimates of the conversion factors for a particular galaxy are then simply given by

$$a_i = m - x_i, \quad i = 1...3.$$
 (F9)

The uncertainty on the factor  $a_i$  depends on the uncertainty on  $m^{\text{opt}}$ , from equation (F8), and the uncertainty on the measurement  $x_i$ , from equation (E1). Since the estimate of *m* depends on the measurements  $x_i$ , there is a non-zero covariance between *m* and  $x_i$ . Allowing for this covariance, the expected uncertainty on  $a_i$  is given by

$$\sigma_{a_i}^2 = \sigma_{\rm m^{opt}}^2 + \sigma_i^2 \left( 1 - \frac{2w_i}{\sum_{i=1}^3 w_i} \right).$$
(F10)

## APPENDIX G: SENSITIVITY OF TRACER TO SFR AND RADIATION FIELD INTENSITY

Fig. G1 shows the observable ratios,  $L_{850}/L'_{CI}$  and  $L_{850}/L'_{CO}$ , as a function of  $T_d$  (left-hand panel) and  $L_{IR}$  (right-hand panel). There is

no significant trend for  $L_{850}/L'_{C1}$  or  $L_{850}/L'_{C0}$  with either  $T_{\rm d}$  or  $L_{\rm IR}$ . There is a noticeable offset to higher  $L_{850}/L'_{C1}$  for the C1<sup>cor</sup> galaxies (pink diamonds), which also have lower  $T_{\rm d}$  and  $L_{\rm IR}$  than the other samples. As these galaxies require large corrections to  $L'_{C1}$  in order to compare to  $L_{850}$ , we cannot be sure if this is a real effect, or just an underestimate of the required correction. A larger sample of low-temperature, low-luminosity galaxies with matched apertures will be required to investigate this.

## **APPENDIX H: TESTS OF ROBUSTNESS**

#### H1 Consistency of parameter estimates

We investigated the consistency of our parameter estimates for the same galaxies when three tracers are used compared to only two. Fig. H1 shows that there is a reasonable correlation between the three-tracer and two-tracer estimates, with only small differences in the sample medians when different numbers of tracers are used. The Xd pair produces the closest match to the method with three pairs (Fig. H1 centre and lower right-hand panels), with no bias and a small scatter. If restricted to choosing only one pair to observe, the best choice seems to be  $L_{850}$  and  $L'_{C1}$ .



Figure G1. Luminosity ratios as a function of luminosity-weighted (peak SED) dust temperature (left-hand panel) and  $L_{IR}$  (right-hand panel). In the top row, the resolved local galaxies from J19 which require aperture correction are shown as pink diamonds. In the bottom row, the galaxies with  $f_{HI} > 1$  are shown as cyan diamonds, after correction following equation (B1). The  $L'_{CI}/L'_{CO}$  ratio has a significant correlation with  $T_d$  and with  $L_{IR}$ , and is shown in Fig. 5 in the main text.



 $\alpha_{CO}$  (daX) 0 CLCO 4000 20 **(px**) 2000 <sup>±</sup>⊻ (ad) 15 2000 15 Ŧ 10 10 1000 1000 5 0 0 4000 -0.2 0.0 . 0.4 4000 -0.2 1000 2000 -0.4 1000 2000 -0.4 0.0 0.4 0.2 0.2  $\Delta \log \kappa_H (daX, ad)$  $\Delta \log \kappa_H (daX, Xd)$ к<sub>н</sub> (**daX)** к<sub>н</sub> (**daX)** 30 25 CI cor CI cor 4 4 25 20 <ci (×10<sup>-5</sup>) (Xa) X<sub>CI</sub>(×10<sup>-5</sup>) (**Xd)** 20 15 15 10 10 5 -0.2 0.0 -0.2 2 3 -0.4 0.2 0.4 2 3 -0.4 0.0 0.2 0.4 1 4 1 4  $X_{CI}(\times 10^{-5})$  (daX)  $X_{CI}(\times 10^{-5})$  (daX)  $\Delta \log X_{CI}(daX, Xa)$  $\Delta \log X_{CI}(daX, Xd)$ Figure H1. Comparison of optimized conversion factors derived using three pairs of tracers, daX (x axes of scatter plots) compared to using just one pair.

The histograms show the offsets the parameters (P)  $\Delta P = \log P_{daX} - \log P_S$ . The filled histogram represents galaxies free from systematic uncertainties in C I fluxes for the J19 sample (the affected C1<sup>cor</sup> galaxies are shown as the line histogram). The median offsets are shown as the grey dotted lines on the histograms (calculated excluding the C1<sup>cor</sup> galaxies) and are very small (0.02, 0.006, 0.04 dex for Xa, Xd, ad, respectively). The offset histograms have a maximum of 0.2 dex with an rms of 0.06–0.08 dex. The scatter plots show the robust galaxies as filled circles, with C1<sup>cor</sup> as open circles. The larger median offset for the daX-ad comparison is apparent as a non-linear trend in the  $\kappa_{\rm H}$  values: for  $\kappa_{\rm H}({\rm daX}) < 2000$ , there is a persistent trend for the ad pair to return a higher  $\kappa_{\rm H}$ than the daX three-pair method. The offset is also present in the  $\alpha_{CO}$  parameter (top left-hand panels) but looks more like a constant offset in the calibration rather than a non-linearity. The CIcor galaxies (line histogram, open circles) are biased in the sense that the daX and one-pair methods produce very different values for the same parameters. The difference is smallest for the Xd sample because both daX and Xd are affected by the same corrections to the CI fluxes. The difference is more pronounced when comparing daX to ad or Xa, the reason being that the uncertain correction only affects the daX results (because the correction is between CI and dust continuum) while the ad and Xa samples do not have the CI-dust continuum pair.

#### H2 Impact of using fixed versus variable $T_{\rm mw}$

CI cor

 $\alpha_{CO}$  (ad)

In this section, we test a different approach to  $T_{\rm mw}$ , one of the main physical dependencies that impacts on the calibration of gas masses.<sup>38</sup> To estimate gas mass from  $L_{850}$ , the mass-weighted dust temperature,  $T_{\rm mw}$ , is required.  $T_{\rm mw}$  has been set to 25 K in previous studies (e.g. Scoville et al. 2014, 2016; Hughes et al. 2017), adding to the uncertainty in gas-mass estimates for individual galaxies. However, as we wish to study trends in the conversion factors, we are concerned about the possible effects of systematic trends in  $T_{\rm mw}$ , since these may affect the resulting behaviour of the conversion factors if ignored.

Having determined empirical relationships between z,  $L_{IR}$ , SED colour ( $L_{IR}/L_{850}$ ), and  $T_{mw}$  in Section 2.3.1, we compare the calibration results using these empirically determined  $T_{\rm mw}$  to the standard assumption of constant  $T_{\rm mw} = 25$  K made in the literature. Fig. H3 shows the impact of using our empirical relations (coloured points), versus keeping  $T_{\rm mw}$  fixed (grey points). Each panel shows one of the affected conversion factors derived from either the ad or Xd samples. The trends with luminosity - visible for our default prescription disappear when a constant  $T_{\rm mw} = 25$  K is used.

The histogram of the offsets in each conversion factor when using the empirical  $T_{\rm mw}$  compared to constant  $T_{\rm mw} = 25$  K (Fig. H2) shows that the choice of  $T_{\rm mw}$  makes no significant difference to the median values of the parameters (<0.015 dex). For individual galaxies, the average uncertainty introduced by using a constant  $T_{\rm mw}$  is 0.046– 0.06 dex (1 $\sigma$ ), with a maximum of ~0.2 dex.

Finally, the difference in the gas-mass estimates,  $M_{\rm H2}$ , when using constant  $T_{\rm mw}$  versus our empirical prescription is shown in Fig. H4. At lower  $L_{IR}$ , a constant  $T_{mw}$  produces lower  $M_{H2}$  compared to our

<sup>&</sup>lt;sup>38</sup>This is not to suggest that  $\alpha_{CO}$  is not dependent on the physical properties of the gas, but being optically thick, this line does not have any simple relationship with anything we can empirically determine. Similarly, we have shown that  $Q_{10}$  is not easy to determine per galaxy, but its range is small enough to have no significant impact on our calibration study.



**Figure H2.** Log difference between the conversion factors ( $X_{CI}$ ,  $\delta_{GDR}$ ,  $\alpha_{CO}$ ,  $\alpha_{850}$ ) using the empirical relation for  $T_{mw}$ , compared to a constant  $T_{mw}$  of 25 K. The median log difference for each conversion factor is shown as a vertical line, all are <0.015 dex, meaning the choice of  $T_{mw}$  for the dust does not have a significant impact on the overall average values derived from this study.



**Figure H3.** Running means of the conversion factors as a function of  $L_{IR}$ . The solid coloured points use the variable  $T_{mw}$ , as described in Section 2.3.1. The grey points represent the same galaxies and the same method but this time with  $T_{mw} = 25$  K. Errors are standard errors on the mean. To better sample the full luminosity range, we have used the ad sample for this analysis, except for the  $X_{CI}$  panel which uses the Xd sample, excluding the CI<sup>cor</sup> galaxies. The important finding is that the trends in conversion factors with  $L_{IR}$  disappear when a constant  $T_{mw}$  is assumed.



**Figure H4.** Difference in inferred  $M_{\rm H2}$  when using the empirical prescription for  $T_{\rm mw}$  (Section 2.3.1) compared to constant  $T_{\rm mw} = 25$  K. The top panel represents gas mass derived for the ad sample; the lower panel uses Xd. The pink triangles represent the C I<sup>cor</sup> galaxies. Using a variable  $T_{\rm mw}$  compared to a constant 25 K produces higher [lower] gas masses at log  $L_{\rm IR} < 11$  [log  $L_{\rm IR} > 12$ ] by up to 0.1 dex. This is not a significant issue given all other uncertainties affecting gas mass estimates.

empirical method, because these galaxies are local discs which tend to have colder diffuse dust temperatures. At higher log  $L_{\rm IR}$ >12, the trend reverses as the diffuse dust temperatures increase to ~30 K.

## APPENDIX I: A ROBUST ORTHOGONAL DISTANCE REGRESSION ALGORITHM

In order to fit the most robust linear model to the data, we have employed an ODR and included intrinsic scatter.<sup>39</sup>

<sup>39</sup>These ideas are outlined in Hogg, Bovy & Lang (2010) and Foreman-Mackey (2017), however both of their Bayesian implementations result in biases in the estimate slope. The biases are quite pronounced when the range sampled by the data is not much larger than the errors on the data, but are significant even when the range sampled is  $\sim 10\sigma$ . The biases also depend on which axis is chosen as the 'true' independent variable and whether the errors are asymmetrical ( $\sigma_x \ll \sigma_y$ , or  $\sigma_x \gg \sigma_y$ ). We found that an ODR which does not use the Bayesian likelihood formalism is the only one which does not have such biases; hence our choice to use it here.

We use the EMCEE MCMC sampler (Foreman-Mackey et al. 2013a) to explore the  $\chi^2$  space and compute robust confidence intervals. Our algorithm results in parameters which are symmetric under transformation of *x* and *y*, allowing us to utilize the full co-variance matrix, including the intrinsic scatter as a third variable.

The MCMC is set up to explore the following likelihood function

$$LnL = -0.5 \sum_{i=1}^{N} (\Delta^2 / \sigma^2 + \ln(\sigma^2 / S_2)), \qquad (I1)$$
$$\Delta = \mathbf{v}.\mathbf{Z} - b \cos(\theta),$$

with **Z** as the data array of *x* and *y* values, *b* as the intercept, and  $\theta$  related to the slope as  $m = \tan(\theta)$ . *v* is a matrix to rotate to find the perpendicular distances, given by  $\mathbf{v} = [-\sin(\theta), \cos(\theta)]$ .

 $\sigma^2 = (\mathbf{S} + \Lambda_{\mathrm{m}}.\mathbf{v}).\mathbf{v},$ 

3.7

where **S** is the co-variance matrix. To include intrinsic scatter in the orthogonal direction, as well as measurement errors into the fitting, we add a term to the co-variance matrix, as suggested in Foreman-Mackey (2017)

$$\Lambda_{\rm m} = \left(\tan(\theta)^2 - \tan(\theta) - \tan(\theta) \ 1.0\right) \times \cos(\theta)^2 \times e^{2\ln(\lambda)}, \tag{I2}$$
  
$$S_2 = (\mathbf{S}.\mathbf{v}).\mathbf{v}.$$

The initial conditions were given by the ordinary least-squares fit parameters for variance in the y direction. The run was checked to ensure adequate burn-in and independence between samples. We used 32 random walkers with 6,000 steps each.

This paper has been typeset from a  $T_{\ensuremath{E}} X/I \!\! \ensuremath{\Delta} T_{\ensuremath{E}} X$  file prepared by the author.