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HI content at cosmic noon – a millimeter-wavelength perspective

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ABSTRACT

In order to understand galaxy growth evolution, it is critical to constrain the evolution of its building block: gas. Mostly comprised by Hydrogen in its neutral (HI) and molecular (H₂) phases, the latter is the one mostly directly associated to star-formation, while the neutral phase is considered the long-term gas reservoir. In this work, we make use of an empirical relation between dust emission at millimeter wavelengths and total gas mass in the inter-stellar medium (M_{HI} plus M_{H2}) in order to retrieve the HI content in galaxies. We assemble an heterogeneous sample of 335 galaxies at 0.01 < z < 6.4 detected in both mm-continuum and carbon monoxide (CO), with special focus on a blindly selected sample to retrieve HI cosmological content when the Universe was $\sim 2-6$ Gyr old (1 < z < 3). We find no significant evolution with redshift of the M_{HI}/M_{H2} ratio, which is about 1-3 (depending on the relation used to estimate M_{HI}). This also shows that M_{H2}-based gas depletion times are underestimated overall by a factor of 2-4. Compared to local Universe HI mass functions, we find that the number density of galaxies with M_{HI} $\gtrsim 10^{10.5}$ M_{\odot} significantly decreased since 8–12 Gyr ago. The specific sample used for this analysis is associated to 20-50% of the total cosmic HI content as estimated via Damped Lyman- α Absorbers. In IR luminous galaxies, HI mass content decreases between $z \sim 2.5$ and $z \sim 1.5$, while H₂ seems to increase. We also show source detection expectations for SKA surveys.

Key words: ISM: abundances – galaxies: ISM – submillimetre: ISM

1 INTRODUCTION

Over the last 25 years it has become increasing clear that the starformation (SF) history in the Universe peaked at about 10 Gyr ago (Lilly et al. 1996; Madau et al. 1998; Hopkins & Beacom 2006; Madau & Dickinson 2014). Nevertheless, it is only in the last 5 years, with the advent of spectral scan surveys conducted with Atacama Large (sub-)Millimeter Array (ALMA; Brown et al. 2004), *Jansky* Very Large Telescope (*JVLA*; Perley et al. 2011), and the NOrthern Extended Millimeter Array (NOEMA; Guilloteau et al. 1992), that the community has identified the driver of this effect in a statistically consistent manner. Namely, the cosmological content of molecular-Hydrogen (H₂) in the Inte-Stellar Medium (ISM) peaks at a similar epoch (Aravena et al. 2016; Decarli et al. 2019, 2020; Riechers et al. 2019; Lenkić et al. 2020).

After the fact, this may come as no surprise since: it is long known that star-formation rate surface density correlates well with total ISM gas – in either neutral or molecular phases — surface density (the so-called Schmidt-Kennicutt, SK, law; Schmidt 1959; Kennicutt 1998);

gas in regions with high SF efficiency is mostly in its molecular phase, while in less SF efficient regions gas is mostly in its neutral phase (HI) (Bigiel et al. 2008); and the SK law holds up to high redshifts (e.g., Bouché et al. 2007).

Nevertheless, both neutral and molecular Hydrogen components in galaxies are difficult to directly detect with increasing redshifts. The former, because of its low transition probability, while the former due to the absence of a permanent dipole moment. Hence, indirect tracers are required if one aims to estimate their content in galaxies up to early cosmic times.

Historically, H_2 has been traced via Carbon Monoxide (CO) emission, with the ground based rotational transition (J:1-0) being the reference tracer. If a higher-J transition is available, population-wide line ratios (R_{JJ-10}) are usually adopted to estimate the ground transition emission (e.g., Carilli & Walter 2013). The H_2 mass can then be obtained from the CO-luminosity by considering a conversion factor (α_{CO10}) that remains a topic of discussion. Today, it is agreed that this factor is highly metallicity-dependent, and that in Solar-like metallicity environments, one can adopt the value found in the Milky-Way (see Dunne et al. 2022, and references therein). However, uncertainties in both R_{JJ-10} and α_{CO10} (among others) can easily build up

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and result in significant uncertainties in the estimated molecular gas mass $(M_{\rm H_2}).$ With the advent of deep observations with ALMA and NOEMA, the community also now considers neutral Carbon (CI) as a molecular gas tracer (Gerin & Phillips 2000; Papadopoulos et al. 2004; Tomassetti et al. 2014), making use of its forbidden transitions $[{\rm CII}^3{\rm P}_1$ $^{-3}$ ${\rm P}_0$ and $^3{\rm P}_2$ $^{-3}$ ${\rm P}_1.$ Despite being fainter than CO low-J transitions, its spectral line energy distributions (SLED) is simpler (only three levels), it is optically thin in most extra-galactic environments, and has fewer excitation mechanisms than CO (see Dunne et al. 2022, and references therein). Nevertheless, this alternative also requires a conversion factor with an associated uncertainty and considerable telescope time.

On the other hand, neutral Hydrogen has only been directly detected up to $z \sim 0.2-0.4$ (Lah et al. 2007; Fernández et al. 2016), and up to z = 1.3 with the aid of strong gravitational lensing (Chakraborty & Roy 2023). Otherwise, at high-redshifts, neutral gas has been mostly traced in absorption (Rao et al. 2006; Braun 2012; Zafar et al. 2013) and trace impact parameters in the 0.1-1 Mpc range, significantly larger than the typical visible galaxy size regardless of cosmic time or reference wavelength (i.e., galaxies at high-redshift are intrinsically smaller, Buitrago et al. (2008), while the largest known spiral galaxy is about 200 kpc in diameter Galaz et al. (2015)). Alternative approaches comprise methods making use of optical continuum or spectroscopic properties (e.g., Zhang et al. 2009; Catinella et al. 2012; Brinchmann et al. 2013; Parkash et al. 2018; Bera et al. 2022), or using [CII] 158 μ m as a HI tracer (Heintz et al. 2021), or combining the SK law with the relation between molecular gas fraction and the mid-plane pressure acting on a galaxy disc (Popping et al. 2015, making use of Bigiel et al. (2008); Blitz & Rosolowsky (2006)).

In this work, we present a millimeter-wavelength-based method to estimate the neutral Hydrogen content in galaxies. In Section 2, we detail the methodological basis of our approach. In Section 3, we detail the sample assembled from the literature considered in our analysis. In Section 4, we report our results including the derived HI cosmological content at $1 \le z \le 3$, while in Section 5 we discuss these results. The conclusions are presented in Section 6. The adopted cosmology refers to the results reported by Planck Collaboration et al. (2020, *Planck18* cosmology henceforth), namely $H_0 = 67.7 \, \mathrm{km \ s^{-1} \ Mpc^{-1}}$ and $\Omega_{\mathrm{M}} = 0.31$ (note that in some figures we also use the Hubble constant dimensionless equivalent $h_{P18} = 0.677$).

2 METHODOLOGY

2.1 Estimating ISM gas mass via dust continuum emission

The last decade has shown the potential of using the mm continuum emission of galaxies to retrieve their molecular-gas content up to very high redshifts (Scoville et al. 2014, 2016, 2017; Magnelli et al. 2020). This is based on the fact that the Rayleight-Jeans tail of the dust thermal emission in galaxies is mostly optically thin, thus being a tracer of the total dust mass (Scoville et al. 2014; Orellana et al. 2017, and references therein). The latter is related to the star-formation activity (both its production and heating; Kennicutt & Evans 2012), which is observed to be related to the molecular gas content (Schmidt 1959; Kennicutt 1998; Bouché et al. 2007; Bigiel et al. 2008). This mm-continuum-based conversion factor (referenced to rest-frame 850 μ m, $\alpha_{850}^{\rm H_2}$) has the great advantage that it requires significantly less telescope time while providing an estimated molecular-gas mass density evolution in the Universe in very good agreement with those based in line observations (Section 4.3).

The underlying assumption of $\alpha_{850}^{H_2}$ is that dust is well mixed with the molecular gas and mostly traces H_2 -dominated regions. However, this scenario is likely only applicable to the luminous-end of the KS-law regime (Bigiel et al. 2008), and it has been shown that the M_{dust} (and as a result 850 μ m luminosity, L_{850} , as a proxy) yields a tighter relation with the total gas mass (Orellana et al. 2017; Casasola et al. 2020), thus including neutral gas (HI). In this work, total gas mass in the ISM refers to:

$$M_{ISM} = \alpha_{heavy} (M_{HI} + M_{H_2})$$
 (1)

where $\alpha_{\rm heavy}=1.36$ is the factor that corrects the gas mass estimate in order to account for chemical elements heavier than Hydrogen in the ISM (Croswell 1996; Carroll & Ostlie 2006), and $\rm M_{H_2}=\alpha_{CO}^{H_2}$ $L'_{\rm CO10}$. Unless otherwise stated, we adopt $\alpha_{\rm CO}^{\rm H_2}=2.9$, which is the value reported by Dunne et al. (2022, $\alpha_{\rm CO}^{\rm H_2}=4.0$), but uncorrected for heavier element fraction.

In order to establish a relation between M_{ISM} and L₈₅₀ we have adopted the sample used in Orellana et al. (2017, and references therein). Briefly, it comprises different types of galaxies found in the local-Universe (z < 0.1) for which reliable HI and CO(1-0) detections are available in the literature. The HI fluxes are global ones, as well as the CO measurements obtained with single-dish facilities. The global dust continuum measurements at 850 µm come from Planck mission and are corrected for: (i) systematic offsets observed when compared with SCUBA-850 observations (Orellana et al. 2017); (ii) spectral shape corrections (a multiplicative factor of 0.887 for a spectral-index of 3; Table 14 in the Explanatory Supplement to the Planck Early Release Compact Source Catalogue¹): and (iii) galactic CO (3-2) contamination (3%; Orellana et al. 2017). For interpretation purposes of the results, it must be emphasized that this "global-measurement" approach means the fluxes of the different components are not fully co-spatial, whereby we do include in this analysis HI emission from regions where no dust or CO are detected.

In Figure 1 we specifically show how L₈₅₀ relates with M_{H₂} (upper left panel), M_{HI} (upper right), and M_{ISM} (lower left). Two type of fits are shown in the panels, a log-log linear relation ($log(L_{850}) =$ $m \log(M_{gas}) + C$; red line and shaded region; referred to as LR method henceforth) and a 1-to-1 ratio $(\log(L_{850})/\log(M_{gas}))$; blue line and shaded region; RT method henceforth). The lower-right panel also shows L_{850} versus M_{ISM} , but where $\alpha_{CO}^{H_2}$ was also left free in the fitting process (note that no priors were used to constrain the value). In each panel, we report the best fits (LR in red, RT in blue, the values in parenthesis shows the fit parameters uncertainties) together with the sample's standard deviation (STD) and median absolute deviation (MAD) in the x- and y-axis directions. For reference, we also show higher-redshift galaxies (yellow stars and square) for which there are also direct detections of HI, CO, and dust continuum (Cybulski et al. 2016; Cortese et al. 2017; Fernández et al. 2016), but were not used in the derived fits.

Quantitatively, based on each fit's resulting population STD and MAD values, it is clear that there is a fitting improvement from left to right, and top to bottom, with overall reductions in STD and MAD of 0.3 dex and 0.1 dex, respectively, for RT. The differences for the LR results are even larger. In the lower-right panel, the reported best-fit value for $\alpha_{\rm CO}^{\rm H_2}$ is 1.21 \pm 0.10. The improvement in the sample spread while using this value is small (< 0.1 dex), but we interpret this reduction in $\alpha_{\rm CO}^{\rm H_2}$ as a balancing of the neutral and molecular regions that L_850 is tracing. In other words, there are galaxies or regions in

https://lambda.gsfc.nasa.gov/data/planck/early_sources/explanatory_supplement.pdf

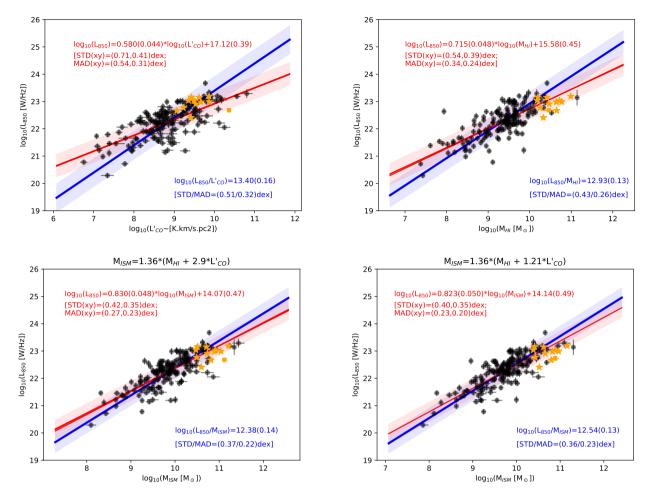


Figure 1. For the sample used in Orellana et al. (2017, black circles), we show how L_{850} relates to $L'_{\rm CO10}$ (as a tracer for $M_{\rm H_2}$; top left-hand side panel), M_{HI} (top right), $M_{\rm ISM}$ (bottom left; Equation 2), or $M^{\rm R}_{\rm ISM}$ (bottom right; Equation 3). The blue line and region show the overall parameter ratio and the data standard deviation (STD) in the log-log space. The red line and region show the best linear fit to the data and its STD in the log-log space. The results are presented in each panel in the bottom right (ratio) and upper left (linear fit), together with the STD and the median absolute deviation (MAD) of the population in the x and y directions. The results from the bottom-right panel are the ones reported in Equations 4 and 5. An higher-redshift sample (Cybulski et al. 2016; Cortese et al. 2017; Fernández et al. 2016) is shown for reference (yellow data points), and intentionally not used in the analysis.

galaxies where M_{H_2} and M_{HI} amount to similar quantities/masses. Considering them together would thus mean doubling the weight of such regions when deriving a fit (i.e., lower-left panel). As such, we interpret this value of $\alpha_{CO}^{H_2}=1.21$ as the limit identifying the regions in galaxies in which L_{850} is tracing a dominant M_{H_2} component of the ISM gas. However, this interpretation needs a more careful analysis, and is deferred to future work. In this manuscript, since we wish to test the implications of using either RT or LT, we adopt $\alpha_{CO}^{H_2}=1.21$ in the adopted RT and LT, but $\alpha_{CO}^{H_2}=2.9$ when estimating total M_{H_2} . As a result, we adopt the following nomenclature throughout the manuscript:

$$M_{ISM} = \alpha_{heavy} (M_{HI} + 2.9 L'_{CO10})$$
 (2)

$$M_{ISM}^{R} = \alpha_{heavy} (M_{HI} + 1.21 L'_{CO10})$$
 (3)

and the following relations:

$$RT = \log\left(\frac{L_{850}[W/Hz]}{M_{ISM}^{R}[M_{\odot}]}\right) = 12.54(\pm 0.13)$$
 (4)

2.2 Rest-frame 850 μ m continuum estimates

We have considered two different approaches in order to estimate the dust continuum flux density at rest-frame 850 μ m (S_{850}^{RF}): (i) modified black body fit to FIR plus (sub-)mm photometry; (ii) power-law fit to (sub-)mm photometry ($S_{\nu} \propto \nu^{\beta}$). The former was pursued by making use of MBB EMCEE² (Foreman-Mackey et al. 2013; Conley 2016) to fit Herschel Space Observatory (Herschel; Pilbratt et al. 2010) photometry together with 0.85–3 mm photometry when available. However, we soon found that this method overestimated S_{850}^{RF} when comparing the fit with and without (sub-)mm photometry. Sometimes, even considering the latter, the Herschel photometry weighted

https://github.com/aconley/mbb_emcee

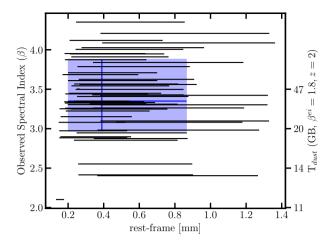


Figure 2. The distribution of the estimated spectral-index in the observed mm spectral range (0.8–3 mm) for 56 galaxies in our sample, for which there are more than one mm-photometry data points. The solid lines show the restframe wavelength range used to estimate the spectral-index. The blue region and errorbar report the median and 16th and 18th percentiles range. The outlier with a low spectral-index is a COLDz galaxy at $z_{\rm spec} = 5.3026$, which is the sole case where the photometry is not tracing the RJ regime. For reference, the right-hand side y-axis shows the dust temperatures reproducing the spectral indices on the left axis. For this exercise, we have adopted a gray (modified black) body model at z = 2 with an emissivity index $\beta^{ei} = 1.8$, estimating the spectral index between 0.6 and 2.6 mm (observed frame; values matching the 16th and 18th percentiles range). We have also considered the effect of a hotter CMB to retrieve observed spectral indices (Section 2.4).

more to the fit, resulting in an overestimated $S_{850}^{\rm RF}.$ As a result, we do not consider those galaxies for which a (sub-)mm detection is not

Approach (ii) was thus the only adopted approach to estimate S_{850}^{RF} . In Figure 2 we show the spectral-index distribution of the 56 sources (47 are from the Birkin et al. (2021) sample; Section 3.2) for which more than one frequency photometry is reported (the minimum and maximum rest-frame are shown with the solid line). The median value of β is $3.35^{+0.53}_{-0.38}$ (MAD=0.26) shown as the blue region and errorbar in the figure. This was the spectral-index adopted in cases where only one (sub-)mm photometry data point is available. We note that it is common to use $\beta = 3.8$, while lower values are associated with estimates based on higher rest-frame frequency values closer to the peak of the dust black-body emission. To test this, we have limited the analysis to the 11 galaxies for which the lowerand upper-wavelength photometry trace rest-frames longer than 300 and $850 \,\mu\text{m}$, respectively. The results remain the same within the uncertainties: $\beta = 3.26^{+0.68}_{-0.32}$

We do a final comparison between the adopted spectral-index flux estimate approach and the case when one normalizes a gray (modified black) body (GB) only to the millimeter spectral range flux estimates (i.e., not considering the FIR spectral range as mentioned above). The spectral energy distribution is thus described with (Hildebrand 1983):

$$I_{\nu}(T_{\rm d}, z) \propto \kappa_{\nu_{\rm rast}} B_{\nu}(T_{\rm d}, z)$$
 (6)

where B_{ν} is the black body function assuming a dust temperature T_d , the dust mass absorption coefficient is $\kappa_{\nu_{\text{rest}}} \propto \nu_{\text{rest}}^{\beta^{ei}}$, and β^{ei} is the dust emissivity index. Following Scoville et al. (2016), we adopt

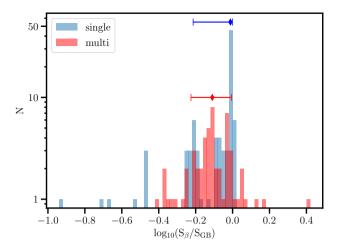


Figure 3. The distribution of the ratio between the power-law and grav-body (GB) predicted dust emission at rest-frame 850 μ m. The blue/red histogram considers the cases when one/two photometry data are available. Note that axis are in logarithmic scale. The errorbars show the 16th, 50th (median), and 84th percentiles.

 $T_d = 25 \,\mathrm{K}$ and $\beta^{ei} = 1.8$. We further consider the effect of a hotter CMB (Section 2.4).

Figure 3 shows the distribution of the ratio between the power-law and GB predicted dust emission at rest-frame 850 µm. Two histograms are displayed for cases when only one or two photometry data are available (blue and red, respectively). The blue histogram (representing most of our cases) very much peaks at a ratio of ~ 1 with a long tail extending to low values (meaning a larger value predicted by the GB assumption), with a median (16th and 84th percentiles) of $\log_{10}(S_B/S_{GB}) = -0.012$ (-0.21 and -0.00040). The same statistics are found to be -0.11 (-0.23 and -0.0058) for the red distribution. The latter is mostly related to the more curved nature of the GB model with respect to the power-law model, and the two-data-point fitting being worst for the GB model (i.e., it always passes between the two points). Having these results, and the fact that a larger dust flux estimate leads to a larger predicted HI content (for a fixed molecular content), we choose to be conservative and adopt the power-law estimate.

2.3 Converting high-J CO transitions to J:1-0

Although CO J:1-0 is the reference transition with which to estimate total M_{H2}, even up to high redshifts (e.g., Hainline et al. 2006; Carilli et al. 2010; Harris et al. 2010; Ivison et al. 2010a, 2011; Riechers et al. 2011), it is a common practice to target a higher-J transition (e.g., $J_{up} = 2, 3, 4$). This owes to the fact that they are brighter, less optically thick, but they are also found at frequencies where the dust continuum emission is also brighter. For that reason, one needs to consider the CO SLEDs to convert from the higher-J transition to the ground one. In this work, we adopt the line luminosity ratios reported for sub-millimeter galaxies in Table 2 in Carilli & Walter (2013) for the samples assembled from Walter et al. (2011), Saintonge et al. (2013), PHIBSS, Birkin et al. (2021), and any other galaxy in other samples at z > 2 (see Section 3 for sample details). Otherwise, we adopt the line luminosity ratios reported for galaxies at 1.0 < z < 1.6in Boogaard et al. (2020). We do this separation following the findings in Boogaard et al. (2020), acknowledging that selection effects tend

to provide samples with more excited CO SLEDs at higher redshifts. We do note that the CO SLEDS reported by Boogaard et al. (2020) at 1.0 < z < 1.6 are in line with those by Daddi et al. (2015), while the SMG CO SLEDs reported by Carilli & Walter (2013) are in line with those by Bothwell et al. (2013) and Birkin et al. (2021), and slightly lower than those by Boogaard et al. (2020) at 2.0 < z < 2.7.

2.4 Corrections to a hotter CMB

Due to the adiabatic expansion of the Universe, the Cosmic Microwave Background (CMB) shows today a black-body spectral energy distribution characteristic of temperature of 2.73 K (Fixsen 2009), while, as we look back in time, the CMB shows a hotter temperature. This results in the molecular gas and dust becoming progressively in thermal equilibrium with the CMB, thus making it harder to detect emission against a brighter/hotter background emission (da Cunha et al. 2013; Zhang et al. 2016). Here, we adopt the correction recipes proposed by da Cunha et al. (2013), namely, we make use of Equations 12 and 18 therein.

2.5 Estimating M_{HI}

In Equations 2 through 5, there are three main observables: S_{850} , $S_{HI}\Delta_{\nu}$, and $S_{CO}\Delta_{\nu}$. Thus, if one detects two of them, the third one can be inferred within the errors associated with both the photometric errors and the intrinsic population scatter shown in Figure 1. Nowadays, with the growing legacy of ALMA and NOEMA surveys, the most common scenario is when a (sub-)mm facility observes both S_{850} and S_{CO} , sometimes in a single observation. As a result, in a statistical sense, one can attempt to derive the HI content in a galaxy population.

We thus combine Equation 2 with Equations 4 and 5 to determine $M_{\rm HI}$:

$$M_{\rm HI}^{\rm RT} = \frac{10^{\log(L_{850}[W/{\rm Hz}]) - 12.54}}{\alpha_{\rm heavy}} - 1.21 \, L'_{\rm CO10} \tag{7}$$

$$M_{\rm HI}^{\rm LR} = \frac{10^{(\log(L_{850}[\rm W/Hz])-14.14)/0.823}}{\alpha_{\rm heavy}} - 1.21~\rm L'_{\rm CO10} \eqno(8)$$

We note that in cases where the derived M_{HI} is negative, we interpret such a result as a galaxy whose ISM gas component is mostly in its molecular phase.

2.6 Error budget

As one can see from the previous sub-sections, there are different steps involved up until when one retrieves the final HI mass estimate. In this section, we summarize the different uncertainties that are being considered in quadrature to retrieve the final error budget associated to the reported HI mass value:

- we have adopted the MAD values associated with the adopted relations (Section 2.1)
- we have considered population STD associated to the reported luminosity ratios between higher-J and J:1-0 transitions from Boogaard et al. (2020), while we have adopted a general error of 0.15 associated to the values reported by Carilli & Walter (2013), since no error is reported therein, but it is nevertheless in line with other works (e.g., Boogaard et al. 2020; Birkin et al. 2021)

- to the reported CO and continuum errors, we have added 10% of the flux in quadrature to account for absolute flux scaling systematics
- for those galaxies with only one photometry data point in the mm spectral range, we have considered the MAD associated to the adopted spectral index.
- in the luminosity functions and cosmological mass density estimates, we account for cosmic variance following Trenti & Stiavelli (2008). We use their calculator³ to compute the error fraction associated to cosmic variance based on redshift range, field size, and number of sources. Based on this exercise, we adopt an error fraction of 0.25 and 0.30 for B21 and ASPECS samples, respectively, to be added in quadrature.

3 SAMPLE SELECTION

As stated in the previous section, in this manuscript, we make use of direct observations of mm-continuum and CO to infer the content of HI gas. As a result, we have assembled from the literature a sample of galaxies that have both a CO emission line detection and (sub-)mm continuum coverage. We mainly choose galaxies with a low-Jup CO transition (typically $J_{up} \leq 3$) in order to have a more reliable conversion to CO J:1-0.

As you will see ahead, specifically for the analysis of the cosmic HI mass content (Section 4.3), we focus only on two samples (Sections 3.1 and 3.2) which provide the simplest selection function.

3.1 ASPECS

We consider galaxies from the ALMA SPECtroscopic Survey in the Hubble Ultra-Deep Field (ASPECS) LP survey (González-López et al. 2020; Walter et al. 2016; Aravena et al. 2016) later followed up at lower frequencies by its VLA equivalent VLASPECS (Riechers et al. 2020). Both programs comprise blind surveys of CO covering an area of 4.6 arcmin². We focus on the CO $J_{up}=1,2,3$ line measurements from Boogaard et al. (2020) for 18 galaxies. For those galaxies, we use the 1.2 mm continuum flux densities from Aravena et al. (2020) and González-López et al. (2020), and the 3 mm flux densities from González-López et al. (2019). We also looked for other sub-mm observations and found a 870 μ m flux density measurement for the galaxy "1mm6" in the Chapin et al. (2011) catalog.

3.2 Birkin et al. (2021)

Birkin et al. (2021) presented ALMA and NOEMA observations of SMGs selected from the ALMA-SCUBA-2 Cosmic Evolution Survey (AS2COSMOS, total area of 1.6 deg²; Simpson et al. (2020)), the ALMA-SCUBA-2 Ultra Deep Survey (AS2UDS, total area of 0.96 deg²; Stach et al. (2019); Dudzevičiūtė et al. (2020)) and the ALMA-LABOCA ECDFS Submillimetre Survey (ALESS, area of 30' × 30'; Hodge et al. (2013); Danielson et al. (2017)). The original catalog provides the $870\mu m$ and 3mm flux densities for 61 galaxies, along with CO emission lines, $J_{up} = 2-5$ for 50 of those galaxies.

For the HI mass density analysis in Section 4.3, we focus on the flux-selected galaxies within the "scan sample" in Birkin et al. (2021): 5 source with $S_{870} = 15 - 20$ mJy in AS2COSMOS and 13 sources with $S_{870} = 8 - 14$ mJy in AS2UDS. Note the complementary flux selection. In additon, we also considered the 13 optical/near-infrared

³ Version v1.03: https://www.ph.unimelb.edu.au/~mtrenti/cvc/CosmicVariance.html

faint galaxies within the "scan sample" and the 30 galaxies within the "Spec - z sample".

3.3 COLDz

The CO Luminosity Density at High-z (COLDz; Pavesi et al. (2018)) survey is a blind survey of CO that covered ~ 9 arcmin² of the COSMOS deep field and ~ 51 arcmin² of the GOODS-North wide field. We found the sub-mm photometry in different surveys. For the galaxies COLDz.GN.31 and COLDz.COS.0 we found the 850µm flux density from the SCUBA-2 Cosmology Legacy Survey Geach et al. (2017); for the galaxies COLDz.GN.14 and COLDz.GN.16 we use the $850\mu m$ flux densities from the SUPER GOODS survey Cowie et al. (2017); for the galaxy COLDz.COS.11 we use the $1100\mu m$ flux density from the AzTEC millimetre survey of the COSMOS field Aretxaga et al. (2011); and finally, using both the Liu et al. (2018) and Jin et al. (2018) catalogs we adopted the $850\mu m$ photometry for the galaxies COLDz.GN.15, COLDz.GN.28, COLDz.COS.6, and both the $850\mu m$ and $1100\mu m$ photometry for the galaxies COLDz.GN.0 COLDz.GN.3,COLDz.COS.1, COLDz.COS.2, COLDz.COS.3. This resulted in 13 selected galaxies.

3.4 PHIBSS

The IRAM Plateau de Bure HIgh-z Blue Sequence Survey (PHIBSS; Tacconi et al. 2013) observed the CO (3-2) line emission for 52 massive, main-sequence star-forming galaxies. These galaxies were chosen from UV/optical/IR surveys to study the molecular gas in galaxies near the cosmic star formation peak of normal galaxies, and were carefully selected to cover a complete M*-SFR plane. They included two redshift bins, at $z \sim 1.1$ and ~ 2.2 . The first bin, z = 1-1.5 includes galaxies from the All-Wavelength Extended Groth Strip International Survey (AEGIS; Davis et al. 2007a), which includes imaging from X-ray to radio and optical spectroscopy. The higher redshift bin, z = 2-2.5, includes galaxies from Erb et al. (2006), Mancini et al. (2011), the BzK sample of Daddi et al. (2010), Magnelli et al. (2012) and the three lensed galaxies cB58 (Baker et al. 2004), "cosmic eye" (Coppin et al. 2007) and "eyelash" (Swinbank et al. 2010, also known as J2135-0102). We found sub-mm observations for some of these galaxies in the following catalogs/surveys. We include $850\mu m$ fluxes from the main catalog of the SCUBA-2 Cosmology Legacy Survey (Geach et al. 2017) for the galaxy EGS13004291 and the fluxes for the catalog of the EGS deep field (Zavala et al. 2017) for the galaxies EGS13011155, EGS13011166, EGS13017707, EGS13018076. For the BzK galaxies we include the 1300μ m fluxes from Magdis et al. (2012). From the Liu et al. (2018) catalog we include the 850μ m flux for the BzK17999 galaxy and both the $850\mu m$ and $1100\mu m$ fluxes for the galaxies BzK12591 and PEPJ123633. For the lensed galaxy "eyelash", we include the $850\mu m$ and $1200\mu m$ fluxes from Ivison et al. (2010b); for the galaxy cB58 we include the 850µm flux from van der Werf et al. (2001); for "cosmic eye" we include the $1200\mu m$ flux from Saintonge et al. (2013); and for the galaxy Q1700-MD94 we include the 1200 µm flux from Henríquez-Brocal et al. (2022). This results in 15 galaxies with sub-mm measurements from the PHIBSS sample.

3.5 PHIBSS2

The PHIBSS2 survey (Freundlich et al. 2019) is an extension of the PHIBSS sample, previously described. In this survey, the CO(2-1) line emission was detected for 60 normal star-forming galaxies at

redshifts z = 0.5 - 0.8. These galaxies were drawn from the North field of the Great Observatories Origins Deep Survey (GOODS-N; Giavalisco et al. 2004), the Cosmic Evolution Survey (COSMOS; Scoville et al. 2007), and the AEGIS survey (Davis et al. 2007b). They were chosen because deep HST, good quality spectroscopy and UV/IR observations are available, while following similar selection criteria as in PHIBSS.

The sub-mm information was gathered from the following surveys/catalogs. For the galaxies XG55 and L14GN022 we used the 850μ m fluxes from the SCUBA-2 Cosmology Legacy Survey (Geach et al. 2017); for the galaxies XC54, XF54 and L14EG008 we used the 850μ m fluxes from the EGS deep field sample of the SCUBA-2 Cosmology Legacy Survey (Zavala et al. 2017); for the galaxy L14GN034 we used the 850μ m flux from the SUPER GOODS survey Cowie et al. (2017). From the Liu et al. (2018) catalog we include the 850μ m and 1100μ m fluxes for the galaxy XA55. This results in 7 galaxies with sub-mm measurements.

3.6 Saintonge et al. (2013)

Saintonge et al. (2013) compiled a sample of 17 lensed galaxies to study dust and gas at the high redshifts of z=1.4-3.1. This goal was achieved combining observations in the FIR with Herschel PACS/SPIRE, and of the CO(3-2) line emission, using the IRAM Plateau de Bure Interferometer (PdBI). Also, they include the 1.2mm continuum photometry from IRAM 30m, Max Planck Millimeter Bolometer array (MAMBO; Kreysa et al. 1998) and Submillimeter Array (SMA) observations. These galaxies are considered to be UV-bright lenses, that show similar SFRs and stellar masses to main-sequence galaxies at high redshift, thus, they are normal star-forming galaxies.

We selected 5 lensed galaxies, that are the detected CO (3-2) line. We discarded the sources cB58, Eye and Eyelash since they are part of the PHIBSS sample.

3.7 Seko et al. (2016)

The sample from Seko et al. (2016) is extracted from a larger one reported in Yabe et al. (2012) where 317 sources selected from optical and NIR surveys (SXDS/UDS; 0.67 deg²) were followed-up spectroscopically, with 71 being assigned a reliable redshift estimate. Of those, 20 were targeted by Seko et al. (2016), with 11 being detected in CO (5-4), of which 5 were detected in mm continuum.

3.8 VALES

The Valparaíso ALMA Line Emission Survey (VALES; Villanueva et al. 2017), used ALMA Band 3 to observe the CO(1-0) line emission in 67 galaxies in the redshift range 0.02 < z < 0.35. This survey targeted galaxies from the Herschel Astrophysical Terahertz Large Area Survey (H-ATLAS; Eales et al. 2010), that covered a total area of ~160 deg² of the sky (Valiante et al. 2016). These galaxies were selected because they are detected near the peak of the SED for a normal, local and dusty star-forming galaxy (Villanueva et al. 2017). The sample have reliable matches with the 6th Sloan Digital Sky Survey data release (SDSS; Adelman-McCarthy et al. 2008). The sample originates from two different selections involving criteria such as SDSS sizes or *Herschel* PACS spectroscopy.

Out of the 67 sources, 49 show a CO (1-0) line detection (>5 σ). For 25 of these galaxies we found the 1 mm continuum using the ALMA archive, from the projects 2016.1.00994.S, 2017.1.01647.S,

Table 1. Analysis Sample Summary — this table reports the different samples we have considered to retrieve HI mass content in this work. The first column reports the aliases used throughout the manuscript. The third to fifth columns report the redshift ranges each sample covers, their surveyed areas, and the number of galaxies in each one.

Sample	Reference	Redshift	Area	N
VALES	Villanueva et al. (2017)	0.01-0.35	160 deg ²	49
PHIBSS	Tacconi et al. (2013)	1.0-2.3	$0.41 \mathrm{deg^2}$	23
PHIBSS2	Freundlich et al. (2019)	0.50-0.78	_	60
S13	Saintonge et al. (2013)	1.4 - 2.7	_	5
W11	Walter et al. (2011)	2.2 - 6.4	_	14
ASPECS	Walter et al. (2016)	0.5 - 2.7	4.6 arcmin ²	20
COLDz	Pavesi et al. (2018)	2.0-5.3	60 arcmin ²	58
S16	Seko et al. (2016)	1.3-1.6	$0.77 \rm deg^2$	11
B21	Birkin et al. (2021)	1.2-4.8	$2.81 \mathrm{deg^2}$	50
			Total	335

and 2017.1.00287.S. These observations are down to a depth of \sim 0.02 mJy/beam, \sim 0.003 mJy/beam and \sim 0.03 mJy/beam, respectively.

3.9 Walter et al. (2011)

Walter et al. (2011) compiled $850\mu m$, CO(3-2) and CI observations for different SMG galaxies. We selected 15 of their catalog galaxies for our sample, while the remainder were discarded due to the uncertainty on the magnification factor.

The sub-mm continuum flux information comes from: Ivison et al. (1998, SMM J02399-0136), Benford et al. (1999, BRI 1335-0417), Ivison et al. (2000, SMM J14011+0252), Barvainis & Ivison (2002, RX J0911+0551), Barvainis & Ivison (2002, F10214, Cloverleaf), Isaak et al. (2002, PSS J2322+1944), Ivison et al. (2002, SMM J163650+4057, SMM J163658+4105), Chapman et al. (2003, SMM J123549+6215), Kneib et al. (2004, SMM J16359+6612), Robson et al. (2004, SDSS J1148+5251), Pope et al. (2006, GN 20, GN 20.2)

3.10 Summary

Table 1 provides an overall perspective of each survey or sample considered here. It includes the number of sources, the range in redshift and, when appropriate, the survey areal size. The names in the first column are the aliases by which we will refer to the samples henceforth. Overall, we have assembled a total of 335 galaxies, selected in different wavelength regimes, from optical continuum or spectroscopy, to near-infrared and (sub-)millimeter. As mentioned already, the samples ASPECS and B21 are the samples providing the simplest selection function hence, these are the two samples used to compute the cosmic HI mass density content in Section 4.3.

The full sample is provided in a master table as supplementary material.

4 RESULTS

4.1 HI mass estimates

We derive non-zero and non-negative M_{HI} estimates for 149 and 145 galaxies while using the RT or LR methods (Section 2), respectively. In Figure 4, we show the M_{HI}^{RT} distribution with redshift. We highlight those galaxies that have more than one photometry data points in the mm spectral range (red dots), and those which are known lensed

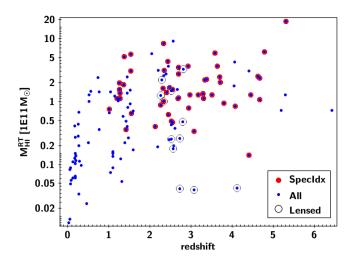


Figure 4. The $M_{\rm HI}^{\rm RT}$ distribution with redshift for 149 with non-zero and non-negative $M_{\rm HI}$ estimates adopting the RT method. The values do not consider the contribution of heavier elements. Galaxies that have more than one photometry data points in the mm spectral range (labelled SpecIdx) are highlighted by red dots. Galaxies that are known lensed galaxies are highlighted by empty black circles. For the latter, the plotted values are corrected for gravitational magnification.

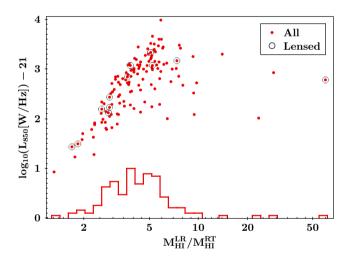


Figure 5. The ratio between the $M_{\rm HI}$ estimates derived with LR and RT methods versus L_{850} . A total of 137 galaxies, for which both methods predict non-zero/negative values, are used in this plot. For reference, the lensed galaxies are highlighted with black circles. The inset histogram is a normalized one to show the distribution of the data set. The distribution shows that LR predicts a median factor of ~4 higher $M_{\rm HI}$ content than the RT method. The y-axis was normalized to 10^{21} for display better visual comparison between the scatter plot and the histogram.

galaxies (empty black circles; magnification-corrected values are displayed). There are no clear deviations between these groups, except for a few lensed galaxies understandably showing lower masses. If one instead plots $M_{\rm HI}^{\rm LR}$ the data points will shift to higher $M_{\rm HI}$ values by a median factor of 4.3 (with a spread of 0.18 dex or a factor of 1.5) as shown in Figure 5 (see also Section 4.2). This is expected given the flatter slope of the LR estimate, that implies a higher $M_{\rm HI}$ component for the same L_{850} and $L_{\rm CO}'$ values with respect to RT.

4.2 ISM gas mass fractions

We now compare the MHI and the MH2 content in our sample. This comparison is shown in Figure 6 for both RT and LR. For reference in the top panel, we mark the equal-content level as a dotted-line, and the median values for RT (dashed blue) and LR (dashed red) methods. The color coding reflects L_{850} . For reference, $L_{850} = 10^{23}$ and 10²⁴ W/Hz are approximately the LIRG and ULIRG thresholds (based on Equation E.5 in Orellana et al. 2017). The middle and bottom panels in Figure 6 show the dependency of the M_{HI}/M_H, ratio with redshift or L_{850} , respectively, for the RT and LR methods (black and red color, respectively). The sample was divided into quantiles, within which the median value is estimated (thick lines). The boxes show where 68% of the population within each quantile falls. These ranges were estimated based on a Bootstrapping analysis, where, for each gas ratio estimate and associated error, we randomly draw 100 new values assuming a log-normal distribution. From these we then retrieve the 16th and 84th percentiles which delimit the boxes. Overall, there is no significant evidence for evolution of the M_{HI}/M_{H2} ratio with redshift or L_{850} , except if one adopts the LR method which may imply higher ratios with increasing L_{850} (red trend in bottom panel). This is not unexpected, since it is at the highest luminosities that the two relations deviate more from each other (Figure 1).

We do note that Chowdhury et al. (2022) reports M_{HI}/M_{H2} ratios of 2–5⁴ for a sample of galaxies with $M^* = 1.03 \pm 0.24 \times 10^{10} M_{\odot}$ at 0.74 < z < 1.45. Although this is in line with the overall value of 2.9 $(-0.42, +0.36 \, \text{dex})$ using LR method, versus $0.8 \, (-0.56, +0.33 \, \text{dex})$ using RT, the galaxy population is different. For those galaxies in our analysis sample at 0.74 < z < 1.45 that have reported stellar mass estimates in the literature, 95% have $\sim M^* > 10^{10.2} \, \mathrm{M}_{\odot}$. Also, the gas mass fractions are sample-selection dependent. Despite the ratio values just mentioned for the analysis sample assembled in this work, the ASPECS and B21 samples used in Section 4.3 show lower values with different deviations in each redshift bin. Namely, the M_{HI}/M_{H2} ratio is 0.9 at $z \sim 1.5$ and 0.5 at $z \sim 2.5$ when using the RT method, and 2.4 and 2.8 when using LR. This goes in line with the fact that these samples are truly dust- and CO-selected (as opposed to other samples that are, e.g., selected in the optical-NIR), hence more likely to yield a larger molecular gas fraction (see next Section 4.3.

4.3 HI mass functions and overall gas density

In this section, we use only the samples from ASPECS and B21 since these are the samples with the simplest selection function, because it only depends on the continuum and CO selections. In the B21 sample, we only consider the sub-sample referred to as "scan sample" (point i-a) therein. We decided not to use the COLDz sample, since the catalogue includes lower fidelity line detections, but the fidelity values are not reported. ASPECS and B21 spectral scans cover similar redshift ranges of interest to this work: 1.01 < z < 1.81 and 2.01 < z < 3.20. The samples are quite complementary, where ASPECS covers better the $M_{\rm H_2} < 6.5 \times 10^{10} \rm M_{\odot}$ regime, and B21 otherwise. We used this $M_{\rm H_2}$ value as threshold above/below which we considered the B21/ASPECS samples. We do this to prevent double counting sources with $M_{\rm H_2}$ values around that threshold while using the V_{max} approach (see next paragraph), especially those above the adopted threshold present in ASPECS since cosmic variance may

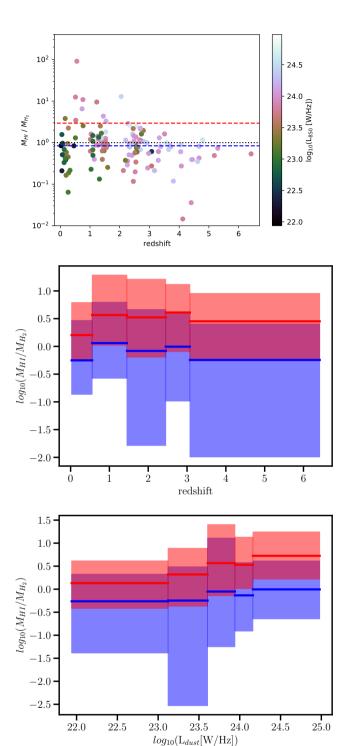


Figure 6. Distribution of $M_{\rm HI}/M_{\rm H_2}$ ratio with redshift and L_{850} . Top panel: The data points show the results adopting RT. The color coding reflects L_{850} . For reference, we mark the equal-content level as a dotted-line, and the median values for RT (dashed blue) and LR (dashed red) methods. *Middle panel*: The dependency of $M_{\rm HI}/M_{\rm H_2}$ ratio on redshift separating the sample into quantiles. The thick lines show the median value in each quantile (red and blue lines for LR and RT methods, respectively). Each box shows where 68% of the population falls ($16^{\rm th}$ and $84^{\rm th}$ percentiles) based on a Bootstrapping analysis (see details in the text). *Bottom panel*: Same as in the middle panel, but with respect to L_{850} .

One detail worth noting is that Chowdhury et al. (2022) estimate the molecular gas content based on a relation dependent on stellar mass and specific star formation rate.

become critical. Nevertheless, Appendix B does show the implications of not adopting this mitigation approach.

In order to determine the volumetric representativeness of each galaxy we adopt the V_{max} method. In order to determine the minimum and maximum redshifts (z_{min} and z_{max}), we consider both the continuum and CO fluxes. The minimum redshift is basically given by the limits of the spectral scans adopted in each survey. To estimate z_{max} , we not only consider the significance of the flux in the detection band and the estimated or adopted spectral-index, but also the significance of the CO detection.

In Figure 7, we show the derived gas mass functions (MFs) for both $M_{\rm H_2}$ (top left-hand panel) and $M_{\rm HI}$ (top right; adopting the RT method, while Appendix A shows the LR method) and compare them with estimates from the literature. All the $M_{\rm H_2}$ measurements from the literature have been converted to the α_{CO} value adopted in this work, and, once more, we do not correct for heavier elements. The agreement with the literature (Decarli et al. 2019; Riechers et al. 2019; Decarli et al. 2020; Lenkić et al. 2020) is noticeable.

The top right-hand panel shows the results for M_{HI} using the RT method. The local-Universe MFs from Zwaan et al. (2005); Martin et al. (2010); Jones et al. (2018) are also displayed for reference (all limited to $M_{HI} < 10^{11} M_{\odot}$). Since the uncertainties in the derived M_{HI} estimate do spread over more than one bin (we adopted a standard bin width of 0.5 dex), we actually distribute the measurement probability per bin assuming a log-normal error distribution since otherwise the massive-end would be underestimated (see Bera et al. 2022, we note that we have also adopted this approach while retrieving the H₂ MFs). Nevertheless, this approach also results in non-zero probability for bins that we believe refer to unrealistic gas masses (see Section 5.4). There is thus the concern that the highmass end may be flatter than reality, while the normalization may be lower than reality. In Appendix C, we show that we do not find a significant trend supporting this expectation. Nevertheless, in line with this exercise, we discard mass bins at $M_{HI} > 10^{12.5}\,M_{\odot}$ for the RT method, and at $M_{HI} > 10^{13} \, M_{\odot}$ for the LR method. We do note that the peaks of distribution may not relate to the intrinsic shape of the functions, but may instead show the mass limit below which each sample is complete at each redshift range. Conservatively, we have thus made the data points more translucent below those thresholds. The thresholds adopted in the H₂ MFs are those from the literature, while those in the H₁ MFs are where one sees the trend inflex (+0.5 and +1 dex higher for RT and LR methods, respectively, with respect to H₂ MFs). Nevertheless, the result seems to point that the main difference in HI content with respect to the local Universe happens in galaxies with $M_{HI} \gtrsim 10^{10.5} \, M_{\odot}$.

The next step is thus to retrieve the cosmological mass density evolution of both dominant ISM gas constituents. In Figure 8 we show measurements from the literature and compare them with our results (left-hand side panel using the RT method, right panel using the LR one). In order to allow for a Cosmology-independent comparison with the literature, what is depicted in the figure is the cosmological mass density:

$$\Omega_{gas} = \frac{\rho_{gas}}{\rho_{crit}} \tag{9}$$

where

$$\rho_{crit} = \frac{3 H_0^2}{8\pi G} \tag{10}$$

is today's Universe critical density. Blue color refers to HI gas, while black/gray refers to H_2 . None of the data points have been corrected for heavier elements.

The assembly of literature measurements for H₂ mass density are also of two types, either from CO detections (Decarli et al. 2020, grey-line limited boxes) or inferred from mm-continuum (Scoville et al. 2017, continuous grey-line limited region). All data have been converted so that the same α_{CO} is adopted (this also applies to the Scoville et al. (2017) results which are based on a relation making use of a CO J:1-0 scaling and $\alpha_{CO} = 6.5$). Our results for H₂ mass density (gray-filled boxes) are quite in agreement with those from the literature (Appendix B shows the results if no M_{H2}-cut is applied between ASPECS and B21 samples). The fact that Scoville et al. (2017) shows a higher cosmological content may be the result of sample incompleteness toward lower M_H, affecting the CO-selected sample. The Scoville et al. (2017) approach was to estimate M_H, with a dependence on redshift, stellar mass, and specific SFR. This allowed the team to account for a more representative population at cosmic noon (see discussions in Sections 3.2 and 10 in Scoville et al. 2017).

The thin blue crosses at the top of Figure 8 are the literature measurements for HI mass density (Zwaan et al. 2005; Lah et al. 2007; Rao et al. 2006; Martin et al. 2010; Braun 2012; Zafar et al. 2013). We note that the measurements above redshift 0.3 are obtained via damped Lyman- α absorption systems, while the local Universe ($z \le 0.24$) measurements are direct detections of the 21 cm line. Our results are shown as thick-line blue error-bars. Within the uncertainties, there is no significant evolution from $z \sim 2.5$ and $z \sim 1.5$. This happens irrespective of the HI-estimate method one adopts, the difference being that RT shows that CO/dust-selected samples recover $23 \pm 12\%$ and $17 \pm 11\%$ of the overall HI mass density at $z \sim 1.5$ and $z \sim 2.5$, respectively, while the LR method points to $61 \pm 27\%$ and $53 \pm 33\%$.

What both methods clearly show is that LIRG-type galaxies have a significant decrease in HI gas content during the same time range (by factors of 0.5 ± 0.4 and 0.4 ± 0.3 for RT and LR methods, respectively; data points shown as blue triangles). Doing the same exercise for the H₂ gas content for the same sub-sample (data points shown as black triangles), we find instead an increase of a factor of 1.4 ± 0.8 , even though with less significance. This trend may explain the drop in SFR density seen for this kind of galaxies since $z \sim 1$ (Le Floc'h et al. 2005; Goto et al. 2011; Magnelli et al. 2011).

4.4 Sources of contamination

Actively accreting super-massive black holes (or Active Galactic Nuclei, AGN) may appear bright at radio wavelengths, to the point that their mm-emission is still dominated by synchrotron emission from the jet (e.g., Messias et al. 2021). If that is the case, then the individual L_{850} estimates will become overestimated, and those of M_{HI} will too as a result (Equations 7 and 8). This may result in a large impact in the cosmic mass density based on a small sample like the one used here.

To test this hypothesis we cross-matched the ASPECS and Birkin et al. (2021) samples with the radio catalogues reported by Simpson et al. (2006, UDS; 1.4 GHz), Kellermann et al. (2008, GOODS-South; 1.4 GHz), Smolčić et al. (2017, COSMOS; 3 GHz). We only find matches with the Birkin et al. (2021) sample of sub-millimeter galaxies (SMGs): a total of 13 sources. One with a counterpart separation of 2 arcsec, and the remainder at < 1 arcsec. Only 3 of the matched sources are used in Section 4.3. Following the approach by Messias et al. (2021), we predict the contribution of the synchrotron emission at rest-frame 850 μ m adopting the radio flux estimates and a spectral index of $\alpha = -0.7$ (where $S_{\nu} \propto \nu^{\alpha}$). We find that 12 sources show an observed-to-predicted flux ratio of $\sim 4-24$ (me-

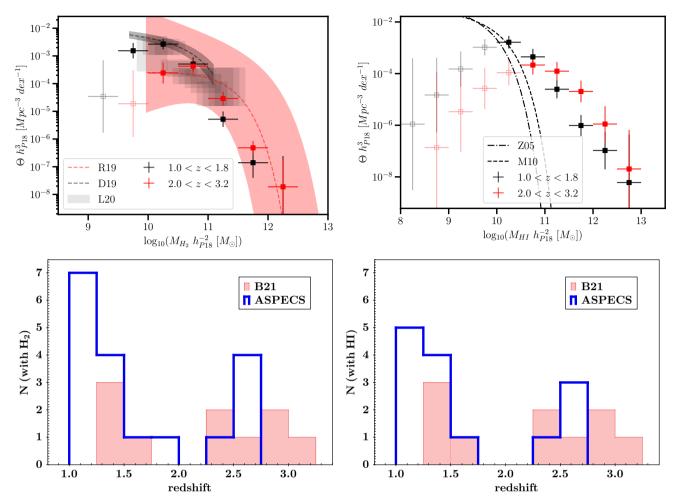


Figure 7. The mass functions (MFs; top row) for $M_{\rm H2}$ (left-hand side panel) and $M_{\rm HI}$ (right panel; results for the RT method; see Appendix A for results with the LR method) obtained with the ASPECS and B21 samples. Black and red data points refer, respectively, to the following redshift bins: 1.01 < z < 1.81, 2.01 < z < 3.20. The literature H_2 MFs come from Decarli et al. (2019, dashed black line and gray shaded region; < z >= 1.43), Lenkić et al. (2020, gray boxes; $z \sim 1-2$), and Riechers et al. (2019, red dashed line and shaded region; 2 < z < 3). The literature $M_{\rm HI}$ MFs are those reported in the local Universe by Zwaan et al. (2005, dashed-dotted line), Martin et al. (2010, dashed line; Jones et al. (2018) results match this one). Translucent data points are indicative of the mass thresholds below which the MFs are believed to be incomplete. The bottom row reports the redshift distributions for ASPECS (blue histogram) and B21 (red shaded region) samples used in this analysis. The left-hand side panel shows the distribution of sources with $M_{\rm H2}$ estimates, and the right panel shows those with positive $M_{\rm HI}$ estimates with the RT method.

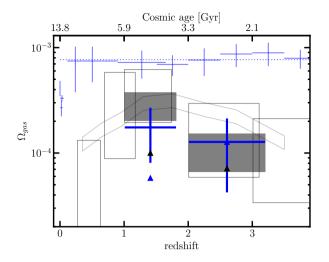
dian of 13), while one (ALESS071.1) shows a ratio of 0.1, which is evidence for its mm emission to be synchrotron-dominated. Curiously enough, the $M_{\rm HI}$ estimate for ALESS071.1 is actually negative (i.e., the ISM gas is mostly in its molecular phase) using either RT or LR methods. Finally, only three sources are also used in Section 4.3 (AS2COS0014.1, AS2UDS627.0, AS2UDS029.0), and for these we find ratios of 16, 9.8, 21 (respectively). Given the order-of-scale difference, we consider these sources not to have a significant synchrotron contribution to their luminosities at rest-frame 850 μm , which also implies that our inferred HI cosmic mass densities are not contaminated (i.e., biased high) due to AGN contamination.

5 DISCUSSION

5.1 The evolution of the HI gas mass density

As mentioned in the previous section, the cosmological HI gas mass density shows no significant evolution for this CO/dust-selected sam-

ple at $1 \le z \le 3$. However, we have reasons to believe that this sample is significantly incomplete in the $z \sim 2.5$ redshift range especially when we compare our H₂ content results with those from Scoville et al. (2017) based on a purely dust-selected sample. If the missing population contributes significantly to the overall HI mass density, then the evolution may be a decreasing one with time from $z \sim 2.5$ to \sim 1.5. This is already apparent if one limits the analysis to the brightest galaxies: $L_{850} > 10^{23}$ W/Hz (for which our sample is expected to be somewhat complete, Figure 9). This LIRG-like sub-sample shows a decrease in HI content of a factor of 2 during this time range, while an increase of a factor of 1.4 in H₂ gas content. This drop in neutral gas inflow may be the reason for the drop in SFR density seen for this kind of galaxies starting ~ 3 Gyr later, i.e., since $z \sim 1$ (Le Floc'h et al. 2005; Goto et al. 2011; Magnelli et al. 2011). The current uncertainties in our analysis are large enough to prevent us from determining how much HI is being converted into H2 between these two redshift bins for this LIRG-like population (currently the errorbars are consistent with all missing HI being converted into H₂).



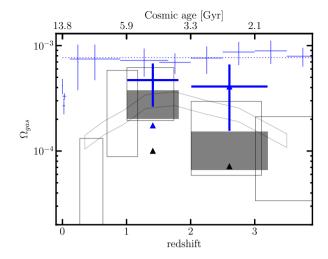


Figure 8. The ISM gas mass cosmological density evolution of its neutral and molecular phases. Left-hand side panel shows the results adopting the RT method, while the right-hand side panel shows those adopting the LR method. Blue error-bars refer to HI gas mass density measurements from the literature, while grey-line limited regions to H_2 gas mass literature estimates (see text for details). The thick line error-bars and grey filled boxes show our results. The blue and black triangles show the HI and H_2 gas mass densities, respectively, for sources with $L_{850} > 10^{23}$ W/Hz (LIRG-type galaxies).

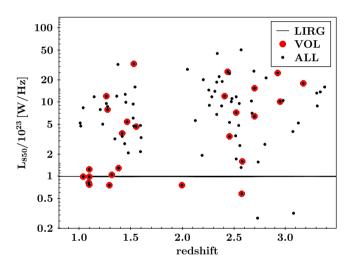


Figure 9. The distribution of L_{850} with redshift for the sample considered in this work, zoomed-in to the redshift range of interest in Section 4.3. The sample used for the cosmic gas density content ("VOL") is highlighted with red dots. For reference, the horizontal line shows the LIRG-like luminosity threshold.

5.2 ISM gas depletion times

In Figure 6, we show that, overall, there is a 1-to-1 (RT) or 3-to-1 (LR) HI-to- H_2 content ratio, with no clear signs of evolution with redshift, except at the highest luminosities (Figure 7). This is thus a hint that the usual depletion times quoted in the literature taking into account only the H_2 content are underestimated by a factor of 2 to 4. However, this is very much sample-selection dependent, since the CO/dust-selected sample used for the cosmological gas content evolution analysis shows median HI-to- H_2 content ratios of 0.9 and 0.5 at $z \sim 1.5$ and ~ 2.5 , respectively, when using the RT method. Nevertheless, this still shows that a significant ISM gas content (at least a third) is not being considered for the estimate of gas depletion times.

For a quantitative example, we make use of the 12 SMGs reported

in Section 4.4 not to have significant AGN contamination in the radio (1.2 < z < 4.8). For these we estimated the SFR adopting the calibration based on the 1.4 GHz continuum emission reported in Murphy et al. (2011) (that assumes the initial mass function from Kroupa 2001):

$$SFR_{1.4 \text{ GHz}}[M_{\odot} \text{ yr}^{-1}] = 6.35 \times 10^{-22} \text{ L}_{1.4 \text{ GHz}}[\text{WHz}^{-1}]$$
 (11)

This retrieves a median SFR of $930\,M_\odot/yr$ (with a minimum-maximum of $\sim\!500{-}2000\,M_\odot/yr$). We find that the median depletion time ($\tau_{SFR}=M_{H_2}/SFR$) considering only $\alpha_{heavy}M_{H_2}$ is $\sim 410\,Myr$ (with a minimum-maximum of $\sim\!260{-}730\,Myr$). If we instead consider the total ISM gas mass (Equation 1) adopting the RT method, the median τ_{SFR} is now $\sim\!880\,Myr$ ($\sim\!350{-}1600\,Myr$), while adopting the LR method, the values become $\tau_{SFR}\sim2700\,Myr$ ($\sim\!710{-}5600\,Myr$). These depletion time values assuming the total ISM gas mass are in line with the predictions from the cosmological hydrodynamic galaxy formation simulation of SMGs reported by Narayanan et al. (2015).

Moreover, in light of the latest findings on Giant Molecular Clouds (GMCs) lifetimes and SF efficiencies (Chevance et al. 2023, and references therein) we would like to raise awareness on the usage of the $\rm M_{H2}/SFR$ ratio to estimate $\tau_{\rm SFR}$ of a system. GMCs in the Milky-Way and other local galaxies are found to have lifetimes of $\tau_{\rm GMC}=10-30\,\rm Myr$ (Feldmann & Gnedin 2011; Murray 2011; Jeffreson & Kruijssen 2018), their SF efficiency is found to be $\epsilon_{\rm SF}=2-10\%$ (Kruijssen et al. 2019; Chevance et al. 2020), while the cloud mass distribution can be described with a power-law of the form dN/dM \propto M c , where -2.5 < c < -1.5 (Murphy et al. 2011; Mok et al. 2020; Chevance et al. 2023).

We have ran a simple exercise to test how the GMC constraints impose a deviation from the simple $\rm M_{H_2}/SFR$ ratio assumption. We first start randomly drawing GMC masses from the cloud mass distribution assuming c=-2 (Mok et al. 2020, and references therein) within a mass range of 10^4 to $10^8~\rm M_{\odot}$ (i.e., GMC scales; Murray 2011; Chevance et al. 2023). In every iteration, we impose that the total cloud mass does not exceed the total mass in $\rm M_{H_2}$, and that ESFR at $\tau_{\rm GMC}$ lifetime scales (i.e., SFR per iteration: SFR $^{\rm tier}_{\rm GMC}=\rm M_{H_2}^{\rm iter}\times\epsilon_{\rm SF}/\tau_{\rm GMC}$) does not exceed the total SFR. The latter condition is to account for the fact that the commonly used

long-wavelength or SED-based SFRs are values integrated over 100 Myr (Table 1 in Kennicutt & Evans 2012), while GMC lifetimes are 3 to 10 times shorter. So, that condition translates to: $SFR_{GMC}^{iter} \le SFR_{tot} * \tau_{GMC}[Myr]/100$. After each iteration, we sum the adopted au_{GMC} to the cumulative of au_{SFR} , and the used up molecular mass $(M_{H_2}^{\text{iter}} \times \epsilon_{SF})$ is reduced from the $M_{H_2}^{\text{tot}}$. The iterations continue until the leftover $M_{H_{\bullet}}^{tot}$ is $\leq 10^6\,M_{\odot}$ (a scale at the level of the mass on a single GMC). This exercise shows that the M_{H_2}/SFR ratio gives similar gas depletion timescales as adopting $\tau_{GMC} = 10$ or 20 Myr and ϵ_{SF} of 5 or 10%, respectively. If one instead adopts the middle expected values for $\tau_{\rm GMC}$ (20 Myr) and $\epsilon_{\rm SF}$ (5%; also the typical value in the Milky Way, Williams & McKee 1997), then one finds that τ_{SFR} doubles with respect to the simple M_{H_2}/SFR ratio. This assumption is roughly the same as when one randomly draws in each iteration values of $\tau_{\rm GMC}$ and $\epsilon_{\rm SF}$ assuming a flat probability distribution. As a result, we suggest the following statistical correction to τ_{SFR} when estimated considering only M_{H_2} :

$$\tau_{\rm SFR} = C_1 * C_2 * \frac{M_{\rm H_2}}{\rm SFR} \tag{12}$$

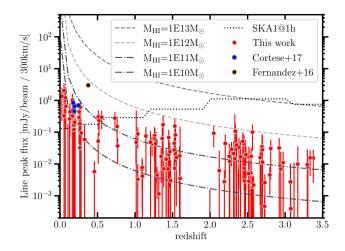
Where C_1 corrects for the existence of a HI reservoir, and C_2 corrects for the $\tau_{\rm GMC}$ and $\epsilon_{\rm SF}$ constraints. A value of $C_1=1$ is equivalent to a molecular gas dominated system, but the population wide results from this work point to $C_1 \gtrsim 2$ (Section 4.2). Note that these values of C_1 assume no extra time delay related to gas transitioning from a neutral to a molecular phase, nor molecular gas dissociation (see section 3.2 in Chevance et al. 2023). As for C_2 , if we adopt average values for $\tau_{\rm GMC}$ and $\epsilon_{\rm SF}$, then $C_2=2$, but once again we note that no dependence was assumed for these two parameters on cloud mass or size (see, for instance, Mok et al. 2020). For that reason, we provide as supplementary material the code used to conduct this exercise in case the reader wishes to build upon the assumptions described above.

5.3 Implications to Square Kilometre Array

As a simple exercise, we make predictions of the HI flux for the sample we have gathered in this work. In order to do so, we use:

$$S_{HI} = \frac{M_{HI} * (1 + z)}{236 * D_1^2 * \Delta v}$$
 (13)

where Δv is adopted to be 300 km/s and a Gaussian-like profile is assumed (even though a double peaked profile is more characteristic of typical galaxies). The line peak fluxes for both RT and LR methods are shown in Figure 10. For reference, we also show the expected fluxes from different MHI values with redshift. The expected SKA 1h on-source integration 3σ sensitivity limit is shown (Braun et al. 2017, 2019). This figure shows that SKA will detect some of the galaxies assembled in this work at least up to $z \sim 0.5$ within 1 h. Above that redshift, it becomes more uncertain, with the both methods implying that, at z > 2, SKA needs to spend at least $\sim 100 \,\mathrm{h}$ to directly detect the bulk of these CO- and continuum-selected galaxies. Based on the obtained HI mass functions (Section 4.3, Figures 7 and A1), Table 2 resumes the expected number of galaxies directly detected by SKA, depending on time on source (ToS = 1, 100 h) and method used to estimate M_{HI} (RT and LR). We separate the estimates in redshift bins given the sensitivity differences in SKA Band 1 at $480-650 \,\text{MHz} \,(1.2 < z < 2.0) \,\text{and} \, 350-480 \,\text{MHz} \,(2.0 < z < 3.1)^5.$



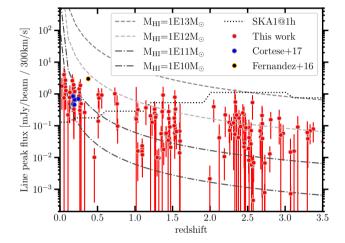


Figure 10. Predicted HI fluxes adopting the RT (top) and LR (bottom) methods for the sample considered in this work (red dots and errorbars). For reference, literature cases with direct HI detections at 0.2 < z < 0.5 are also shown (Cortese et al. 2017; Fernández et al. 2016), as well as expected fluxes for different $M_{\rm HI}$ contents with redshift ($\log(M_{\rm HI}) = 10$ –13). The 1h on-source integration 3σ sensitivity of SKA1 is shown as a dotted line.

We do not present expected numbers for the 2.0 < z < 3.1 case with ToS = 1 h, because the gas mass limit is regarded as unrealistic (see Section 5.4). Note that ToS = 100 h limits the direct detections to $M_{\rm HI} \gtrsim 10^{11} \, \rm M_{\odot}$, where we expect the analysis to be complete. Nevertheless, this sample is still limited by its CO- and continuum-detection nature, hence these numbers should be considered lower limits. Having this, large programs or stacking analysis will be required to characterize the star-forming population at these cosmic times.

5.4 Reliability of RT and LR methods

As shown in Figure 1, the dust-to-ISM gas calibration used local-Universe galaxies mostly with ISM gas masses below $10^{11}~M_{\odot}$, while some of the estimated M_{HI} values for the high-redshift sample reach

⁵ SKA is expected to cover each of these frequency ranges in a single observation, so the ToS value is per frequency range reported.

 $^{^6}$ For completeness, we refer that the results were nevertheless consistent with zero: $0^{+1.2}$ and $0.55^{+4.6}_{-0.42}$ for the RT and LR methods, respectively.

Scenario		redshift	Method		Mass Limit
ToS	Δu		RT	LR	
h	[MHz]		$[deg^{-2}]$		$[\log(M_{\odot})]$
1	480–650	1.2-2.0	$0.5^{+4.2}_{-0.38}$	$3.0^{+5.4}_{-1.4}$	12
	350-480	2.0-3.1	_	_	13
100	480-650	1.2-2.0	120^{+140}_{-62}	960^{+1200}_{-520}	11
	350-480	2.0-3.1	$7.9^{+31}_{-6.2}$	160^{+140}_{-72}	12

Table 2. Expected number of galaxies per \deg^2 at 1 < z < 3 directly detected by SKA based on the obtained HI mass functions (Section 4.3, Figures 7 and A1). We show the numbers for different times on source (ToS) and adopting either RT or LR to estimate M_{HI} . Note that $100\,h$ ToS limits the direct detections to $M_{HI} \gtrsim 10^{11}\,M_\odot$. We do not present expected numbers for the 2.0 < z < 3.1 case with ToS = 1 h, because the gas mass limit is regarded as unrealistic (see Section 5.4).

one order of magnitude higher. Although baryonic masses reaching or in excess of $10^{12}\,M_\odot$ may be expected for extreme cases (see, for instance, the stellar mass functions at similar redshifts in Weaver et al. 2023), we pursue an assessment of the credibility of each method. In this section, we put our results into perspective with respect to physical constrains that may potentially be regarded as strict.

For instance, the galaxy with the largest estimated HI mass content in the 1.0 < z < 3.2 range (ALESS006.1 at z_{spec} = 2.3368) shows M_{HI}^{RT} = (8.4 ± 6.3) × 10¹¹ M_{\odot} or M_{HI}^{LR} = (4.4 ± 3.2) × 10¹² M_{\odot} . The extreme M_{HI} value may be thought as associated with the low estimated continuum spectral-index ($\alpha = 2.41$) resulting in an overestimate of the dust continuum, but the 3 mm continuum detection actually directly traces the reference rest-frame wavelength 850 μ m. Also, from B21 and Section 4.4, there is no evidence for AGN contamination. Compared to its $M_{H_2} = (3.2 \pm 1.3) \times 10^{11} \,\mathrm{M}_{\odot}$, and to the values reported by B21 for stellar and dust masses of $\log(M^* \, [M_{\odot}]) = 11.0 \pm 0.5$ and $\log(M_{dust} \, [M_{\odot}]) = 9.31^{+0.04}_{-0.14}, \; respectively.$ spectively, we find HI fractions of $67^{+20}_{-46}\%$ and $91^{+6}_{-30}\%$ for the RT and LR methods, respectively. Although the uncertainties are large and significant larger HI fractions are expected with increasing redshift (Chowdhury et al. 2022), the RT method estimate is regarded as more realistic, since the high HI fraction estimates by the LR method reach the expected properties of dwarf galaxies (Huang et al. 2012) or very high redshift samples (Heintz et al. 2022), none of which are similar to ALESS006.1.

Another test that one can pursue is by making use of the tight relation between HI mass and the diameter of the HI disc (Broeils & Rhee 1997). In Wang et al. (2016) it is found to be $\log(D_{HI}) = 0.506 \log(M_{HI} - 3.293)$ with a scatter of 0.06 dex. Directly applying this relation to the case of ALESS006.1 would imply a HI size of $550 \,\mathrm{kpc}$ (assuming $M_{\mathrm{HI}}^{\mathrm{RT}}$). Nevertheless, it is known that galaxies at high redshift are intrinsically smaller (Trujillo et al. 2007; Buitrago et al. 2008; Buitrago & Trujillo 2023; van der Wel et al. 2014, 2023). At the redshift of ALESS006.1, late-type galaxies are about 2.5 times smaller, while massive spheroids and/or quiescent galaxies can be factors of 4 - 7 smaller. These values thus imply sizes for ALESS006.1 of ~220 kpc down to ~80 kpc. These values are still significantly larger than the observed stellar sizes at high redshifts (e.g., < 20 - 30 kpc at $z \sim 1$; Buitrago & Trujillo 2023)), which is not surprising, but we must recall that the starting point of this work is a CO- and dust-based empirical relation, components which are expected to be comparable in size to the stellar component. However, we do note that if one considers that the molecular gas mass in ALESS006.1 ($M_{H_2} = (3.2 \pm 1.3) \times 10^{11} \, M_{\odot}$) was totally in its HI phase at a given point in the past, the minimum size assuming the above reasoning would still be $\sim 50\,\mathrm{kpc}$. At this point, we avoid addressing which relation does not hold at these redshifts — the HI mass-size relation or the RT method — but it is clear that the LR method implies much more extreme results, some deemed unrealistic.

Given the points presented in this section and results reported throughout the manuscript, we suggest the usage of the RT method over the LR one, and that the different sources of errors are taken into account (Section 2.6) in order to retrieve a realistic precision on the estimated HI mass based on the RT method.

6 CONCLUSIONS

In this work, we retrieve the atomic Hydrogen (HI) cosmological mass content at cosmic noon (1.0 < z < 3.2). We start by calibrating an empirical relation between millimeter continuum emission at rest-frame 850 μ m in galaxies and their inter-stellar medium (ISM) gas mass (M_{ISM}) with a local-Universe sample (z < 0.1; see Section 2). Such relation has two flavors that we address throughout the manuscript: the so-called RT method assumes a simple ratio between the mm-continuum luminosity and M_{ISM}, while the LR method assumes a log-log linear relation between the two. We then continue with applying this relation to a heterogeneous sample retrieved from the literature (0.01 < z < 6.4; Section 3). With a special focus on a sub-set of galaxies selected in the millimeter wavelength range, namely being detected in CO ($J_{up} = 1, 2, 3$) and in continuum at 0.8– 3 mm (observed frame), we also derive the HI cosmological mass content at cosmic noon (1.0 < z < 3.2; Section 4.3). Based on these results, we also present implications to the Square Kilometre Array (SKA). Overall, we conclude the following:

- Based on the results and putting these into perspective with the literature, we give preference to the RT method over the LR (Section 5.4);
- On a galaxy population wide view, we find no significant evolution on the atomic to molecular gas mass ratio, but specific sample selections may show differences with cosmic time (Section 4.2);
- Based on this finding and in light of recent findings regarding the properties of Giant Molecular Clouds, we show that depletion times purely based in molecular gas content are underestimated by at least a factor of 2, but potentially by 4 (Section 5.2).
- We find no significant evolution in the range 1.0 < z < 3.2 of the HI cosmological gas content in (sub-)mm-selected galaxies, but this is a result of the sample selection being limited to more luminous/massive galaxies in the high-redshift range targeted here (Section 4.3):
- We find tentative evidence for a decrement in HI gas mass in luminous infrared galaxies (Section 4.3);
- Finally, we find that stacking analysis or large programs conducted with the SKA are required to study the bulk of the galaxy population referred in this manuscript where a 100 h on-source project would directly detect \sim 120 sources per square degree at 1.0 < z < 2.0, and an order of scale less at 2.0 < z < 3.1 (Section 5.3).

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This paper makes of the following use **ALMA** ADS/JAO.ALMA#2016.1.00994.S, data: ADS/JAO.ALMA#2017.1.01647.S,

ADS/JAO.ALMA#2017.1.00287.S. ALMA is a partnership of ESO (representing its member states), NSF (USA) and NINS (Japan), together with NRC (Canada), MOST and ASIAA (Taiwan), and KASI (Republic of Korea), in cooperation with the Republic of Chile. The Joint ALMA Observatory is operated by ESO, AUI/NRAO and NAOJ.

DATA AVAILABILITY

The information and data used in this manuscript is described in Section 3. Specifically in Table 1 we present the summary of the original works where the sample was retrieved from. Whenever needed, we also mention archival data used to complete the required information to pursue our work.

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APPENDIX A: RESULTS WITH LR METHOD

In this section, we complete Figure 7 by showing the HI mass functions using the LR method in Figure A1.

APPENDIX B: RESULTS WITHOUT THE M_{H2}-CUT

As mentioned in Section 4.3, we have adopted a cut in $M_{\rm H_2}$ between samples ASPECS and B21 so to guarantee complementary between the two. Nevertheless, here we present the results (Figures B1 and B2) when such a cut is not applied. The main differences to highlight are: the expected increase in cosmic gas content in both gas phases (most noticeably in $M_{\rm H_2}$) and redshift bins; the LR method implies that, within the errors bars, the samples extracted from ASPECS and B21 already recover the $M_{\rm HI}$ content estimated from DLA

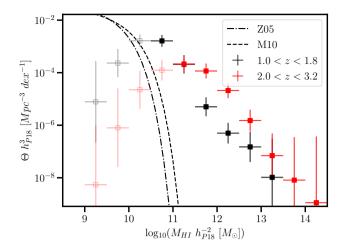


Figure A1. The same as in the top right-hand side panel of Figure 7, but adopting the LR method.

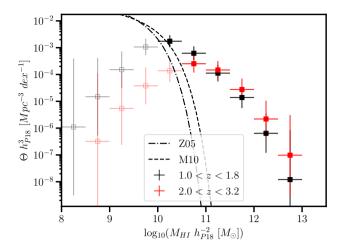
systems analysis; the sample at $z \sim 2.5$ still shows evidence for incompleteness with respect to the S17 approach; the MHI content from the LIRG-like population is constant, but the M_{H_2} content still increases.

APPENDIX C: RESULTS WITHOUT CONSIDERING HI MASS UNCERTAINTY

Given the large uncertainties associated to the HI mass estimate on an individual basis, we pursued a statistical analysis where the probability of a source to fall in a given mass bin was estimated adopting a log-normal probability distribution function (PDF) described by the most probably value and the associated error. In this section, we show the differences between this approach and the case if we would not consider the gas mass PDF, whereby a galaxy is associated to the bin comprising its most probable mass estimate value. In order to build such mass functions (MFs) with our reduced galaxy sample, we followed the standard strategy that works in the literature have been using to build CO luminosity functions (Decarli et al. 2019, 2020; Riechers et al. 2019, 2020; Lenkić et al. 2020). Briefly, we use mass bins 0.5 dex-wide separated by 0.2 dex steps. Subsequent steps are not independent, but our goal here is to find significant deviations from the results when the mass PDF is considered.

In Figure C1, we directly compare both strategies at 1.0 < z < 1.8(left-hand side column) and 2.0 < z < 3.2 (right column), and also for the RT (top row) and LR method (bottom row). For this exercise, we do not consider the cosmic variance error budget since we are comparing methods, while the sample is the same. As described in the main text, the expected trend resulting from using the gas mass PDF with respect to using single value is to a lower normalization and a flatter massive end. Although we see the tendency for the PDF analysis to be lower at the low-mass end, there is still agreement within the 1σ error bars, and we also see agreement at the massive end, especially in the lower-redshift range. Moreover, the light-end may be affected by lower-significance estimates that may be boosted by noise bias, and the PDF approach may be reducing this effect. However, we do not have the tools to confirm this point.

Based on this exercise, however, one can observe that the most extreme bins at the massive end do not trace ranges covered by



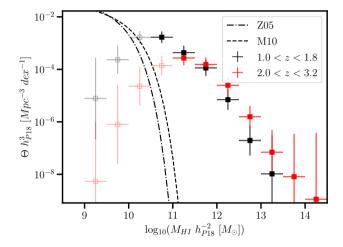
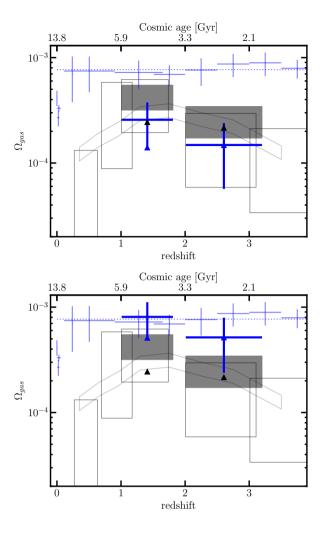


Figure B1. The same as in the top right-hand side panel of Figure 7, but not applying the cut in M_{H2}. Top panel considers the RT method, while the bottom one the LR method.

the most probable values estimated to this sample. As a result, we intentionally remove them from the figures displayed in the main text. Specifically, we discard mass bins at $M_{HI} > 10^{12.5}\,M_{\odot}$ for the RT method, and at $M_{HI} > 10^{13}\,M_{\odot}$ for the LR method.

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 $\label{eq:Figure B2.} \textbf{Figure B2.} \ \text{The same as in Figure 8, but not applying the cut in } M_{H_2}. \ \text{Top panel considers the RT method, while the bottom one the LR method.}$

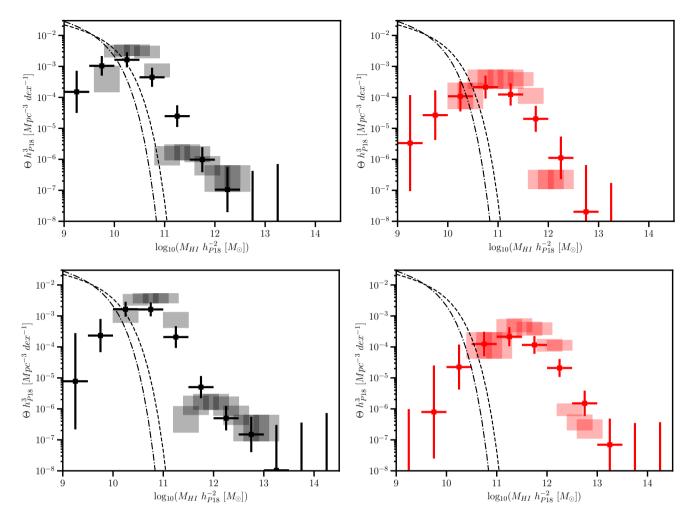


Figure C1. This figure compares the MFs when one distributes the source probability over different bins (Section 4.3) versus when one adopts only the highest probability value. The MFs adopting the latter approach were obtained by estimating the $\Sigma_t V_{max}^i$ in 0.5 dex wide bins, at each step of 0.2 dex (following the usual approach in the literature to build CO LFs; e.g., Decarli et al. 2019, 2020; Riechers et al. 2019, 2020). The top row shows the results adopting the RT method, while the bottom row those with LR. The left-hand side column shows the results for the 1.0 < z < 1.8 range, while the right shows those for the 2.0 < z < 3.2 range. The local HI mas functions are displayed for reference (Zwaan et al. 2005; Martin et al. 2010).