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On the dust properties of high-redshift molecular clouds and the connection to the 2175 Å extinction bump

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ABSTRACT

We present a study of the extinction and depletion-derived dust properties of gamma-ray burst (GRB) absorbers at 1 < z < 3 showing the presence of neutral atomic-carbon (C I). By modelling their parametric extinction laws, we discover a broad range of dust models characterizing the GRB C I absorption systems. In addition to the already well-established correlation between the amount of C I and visual extinction, A_V, we also observe a correlation with the total-to-selective reddening, R_V. All three quantities are also found to be connected to the presence and strength of the 2175 Å dust extinction feature. While the amount of C I is found to be correlated with the SED-derived dust properties, we do not find any evidence for a connection with the depletion-derived dust content as measured from [Zn/Fe] and N(Fe)_{dust}. To reconcile this, we discuss a scenario where the observed extinction is dominated by the composition of dust particles confined in the molecular gas-phase of the interstellar medium (ISM). We argue that since the depletion level trace non-carbonaceous dust in the ISM, the observed extinction in GRB C I absorbers is primarily produced by carbon-rich dust in the molecular cloud and is therefore only observable in the extinction curves and not in the depletion patterns. This also indicates that the 2175 Å dust extinction feature is produced by dust and molecules in the cold and molecular gas-phase. This scenario provides a possible resolution to the discrepancy between the depletion- and SED-derived amounts of dust in high-z absorbers.

Key words: gamma-ray burst: general – ISM: abundances – dust, extinction – galaxies: high-redshift – galaxies: ISM.

1 INTRODUCTION

One of the key constituents of galaxy formation and chemical evolution is the interplay and connection between dust, gas, and metals in the interstellar medium (ISM). Since stars form in molecular clouds, identifying the cold neutral gas-phase in the ISM of galaxies in the early Universe will provide imperative clues to the onset of star formation. A powerful probe of the ISM in high-redshift galaxies are damped Lyman-α absorbers (DLAs), which can be observed towards background quasars (Wolfe, Gawiser & Prochaska 2005) or in gamma-ray burst (GRB) host galaxies (Jakobsson et al. 2006; Fynbo et al. 2009).

Detailed information of the gas, metal, and dust content of DLAs can be derived from absorption-line analyses or from the extinguished spectral energy distribution (SED). The optical afterglows of GRBs provide a unique tool to study the absolute extinction curves of their host galaxies because of their simple power-law

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spectra (e.g. Schady et al. 2010; Greiner et al. 2011; Zafar et al. 2011; Covino et al. 2013), which is more difficult to constrain in quasar sightlines due to the more unknown variation in the underlying spectrum (but see e.g. Vladilo et al. 2006; Jiang et al. 2011; Ledoux et al. 2015; Kro­gager et al. 2016; Fynbo et al. 2017; Noterdaeme et al. 2017; Heintz et al. 2018a). In addition, the extinction curves provide information about the dust content, composition and grain size distribution compared to the attenuation of the GRB host (which can be studied in emission after the afterglow has faded, e.g. Corre et al. 2018), since the latter effect is a combination of reprocessed photons, dust-star geometry and radiative transfer effects (Narayanan et al. 2018).

An alternative tracer of dust is the depletion of refractory elements such as silicon and iron from the gas-phase of the ISM (Savaglio, Fall & Fiore 2003; Jenkins 2009; De Cia et al. 2016). It is expected that the amount of dust derived from the SED, typically represented as the visual extinction, A_V, should then scale with the elements in the dust-phase (Vladilo et al. 2006; De Cia et al. 2013). However, there appears to be a discrepancy between the SED- and depletion-derived dust properties of GRB host absorption systems (Savaglio & Fall 2004; D’Elia et al. 2014; Wiseman et al. 2017). This inconsistency might be due to the fact that iron (typically used to represent the dust-phase and argued to be the most reliable depletion measure; De Cia 2018) does not trace most of the dust mass that will otherwise impact the inferred extinction (Zafar & Watson 2013; Dwek 2016). Another possibility is that the SED-derived visual extinction is underestimated if the typical grain size distribution is large, resulting in grey extinction (Savaglio et al. 2003). We note though that Bolmer et al. (2018) found that this tension is relieved at z \geq 4, at least in H_2-bearing GRB absorbers, and that Zafar & Mottler (2019) showed that A_V is related to the total column density of refractory elements.

The cause of this putative discrepancy between the dust content inferred from the extinguished afterglow SED and the gas-phase depletion levels might also be related to a too simplistic view of the location of the dust. Recent studies of GRB and quasar absorbers have for instance found that the visual extinction, A_V, and the strength of the 2175Å extinction bump feature is correlated with the amount of neutral atomic-carbon (C_I; Zafar et al. 2012; Ledoux et al. 2015; Ma et al. 2018; Heintz et al. 2019). Absorption features from C_I are often found to be coincident with H_2-bearing GRB absorbers, and that Zafar & Mottler (2019) showed that A_V is related to the total column density of refractory elements.

Carbonaceous material could produce the rare 2175Å bump (Stecher & Donn 1965; Weingartner & Draine 2001), since carbonaceous grain growth requires neutral atomic-carbon and molecules in the ISM (Henning & Salama 1998). The 2175Å dust extinction feature is ubiquitously observed locally in the MW (Fitzpatrick & Massa 2007) and in the LMC (Gordon et al. 2003), but is mostly elusive in the SMC and in extragalactic sightlines (Gordon et al. 2003; Noll et al. 2009; Zafar et al. 2012; Ledoux et al. 2015; Zeimann et al. 2015; Ma et al. 2018). The strength and width of the characteristic feature vary with the surrounding environment but its central wavelength has been found to remain invariant (Fitzpatrick & Massa 1986, 2007). Early studies have suggested that graphite or polycyclic aromatic hydrocarbons (PAHs) are responsible for the 2175Å dust extinction feature (Stecher & Donn 1965; Draine & Lee 1984; Léger et al. 1989; Blanco, Fonti & Orofino 1996; Li & Greenberg 1997; Henning & Salama 1998; Weingartner & Draine 2001; Draine 2003). More recently, Mishra & Li (2017) reported that the strength of the 2175Å dust extinction feature is related to the amount of carbon in the dust-phase for a sample of 46 Galactic sightlines, further supporting carbonaceous dust grains as the carriers producing the 2175Å dust bump.

In this paper, we study a sample of C_I absorbers identified by Heintz et al. (2019, hereafter Paper I) in GRB sightlines, with the aim of investigating the mineralogy, dust composition and grain size distribution of the cold and molecular gas-phase in these high-z absorbers. Here, for the first time, we attempt to distinguish the different contributions of various elements that could be the carrier of the 2175Å dust bump and affect the shape of the extinction curve at high redshifts. This sample of GRB C_I absorbers has the advantage that for example the total-to-selective reddening parameter, R_V, can be well constrained compared to similar systems in quasar sightlines. Spectroscopy of the optical afterglows of the GRBs in this sample (at a redshift range of 1 < z < 3) thus provides a unique way to study the dust content of molecular clouds in star-forming galaxies at the peak of cosmic star formation. We have acquired elemental abundances and derived extinction curves for each of the GRB C_I absorbers entering our sample, and combined with previous literature measurements we examine the distinct dust phases of this unique sample of GRB C_I absorbers.

We have structured the paper as follows. In Section 2, we present the GRB C_I absorber sample, including their basic individual properties such as redshift, gas-phase abundances and C_I content. In Section 3, we describe our approach of computing the dust extinction laws and we provide our results in Section 4. We discuss a specific scenario on the location of the dust in the sightline to the GRB C_I absorbers in Section 5, and conclude on our work in Section 6. Throughout the paper, errors denote the 1σ confidence level. We assume a standard flat cosmology with H_0 = 67.8 km s^{-1} Mpc^{-1}, Ω_m = 0.308, and Ω_{\Lambda} = 0.692 (Planck Collaboration XIII 2016). Gas-phase abundances are expressed relative to the Solar abundance values from Asplund et al. (2009), where [X/Y] = log(N(X)/N(Y)) − log(N(X)_\odot/N(Y)_\odot), following the recommendations of Lodders, Palme & Gail (2009) on whether to use the photospheric or meteoritic values (or their average).

## 2 Sample

The identification of GRB C_I absorbers in Paper I was based on bursts observed as part of the GRB afterglow sample by Fynbo et al. (2009) and the VLT/X-shooter GRB (XS-GRB) afterglow legacy survey (Selsing et al. 2019). Neutral atomic-carbon was detected in 10 GRB optical afterglow spectra obtained with low- to medium-resolution spectrographs (R = 1000–10,000). The detection rate was found to be ≈25 percent in the full statistical sample of GRBs compiled in Paper I at a completeness limit of W_r(λ, 1560) = 0.2 Å. An overview of the GRBs with C_I identified in absorption is provided in Table 1, also including those bursts not entering the statistical sample from Paper I. The only C_I-bearing GRB absorber from Paper I not included in this work is GRB 060210. This burst was excluded from our analysis because the X-ray spectrum from the Swift/XRT appears to be affected by X-ray flaring at the time when the optical spectrum was obtained. It is therefore not possible to constrain the intrinsic slope of the SED of this GRB, required to quantify the dust-extinction model. We note that Curran et al. (2007) also find an offset between the X-ray and optical slopes based on their R- and I-band photometric data.

For all the nine GRB C_I systems studied here, we provide the gas-phase abundances of H_I, ZnIII, SiII, and FeII in Table 2, where
available from the literature. In the Appendix, an individual note on each GRB is given with references to where the literature values were obtained from. Based on the gas-phase abundances, we also list the derived metallicity, [Zn/H], the dust depletion, [Zn/Fe], and the dust-phase iron column density, \(N_{\text{Fe}}^{\text{dust}}\), and \(N_{\text{Si}}^{\text{dust}}\) for each GRB where possible. The actual metallicity of the C I neutral hydrogen column density ranges from \(\log N_{\text{H}} = 20.7\) to 21.5, where the 2175 Å extinction bump is clearly detected. Dust in GRB C I absorbers is found to be an ideal tracer of dust in the ISM (De Cia et al. 2018). The dust-phase iron column density, \(N_{\text{Fe}}^{\text{dust}}\), is in most cases also found to be correlated with the visual extinction, \(A_V\), derived from the SED of the optical to X-ray afterglow spectra (Vladilo et al. 2006; De Cia et al. 2013), further supporting the use of depletion patterns as a tracer of dust in the ISM.

In Paper I, we found that C I was exclusively identified in GRB host absorption systems with large metal column densities (\(\log N(X) = \log N(H) + [X/H] > 20.7\)), dust-phase iron column densities of \(\log N_{\text{Fe}}^{\text{dust}} \geq 16.2\), and constitutes the most dust-reddened population of GRBs at all redshifts. This is not unexpected, since a large metal and dust content is required to shield the neutral gas from the ambient galactic UV field. From Table 2 we find that the neutral hydrogen column density ranges from \(\log N_{\text{H}} = 21.5\)–22.7 and that the GRB C I absorbers have abundances between one-tenth of solar to solar values. We refer the reader to Paper I for a detailed discussion of the metal content of the GRB C I systems, where we also compared them to C I-bearing quasar absorbers. Here we focus on the SED-derived dust properties of the systems, which we derive for each individual burst (combined with literature measurements), and investigate how this relates to the depletion-derived dust properties in this type of cold gas absorbers. While this can in principle also be studied in quasar absorbers, it is typically much easier to constrain the specific dust-extinction models in sightlines to GRBs since their intrinsic spectra are simple, smooth power laws.

3 SED ANALYSIS

To model the parametric extinction laws of each of the individual GRBs in our sample, we follow the same procedure as Zafar et al. (2018a). We refer to their work for the explicit details of the fitting procedure, and only briefly summarize the methodology below. Zafar et al. (2018a,c) already determined the extinction curve parameters for the GRBs 120119A, 120815A, 121024A, and 180325A entering our sample, so here we provide only the best fits for GRBs 061121, 070802, 080605, 080607, and 150403A, using the same dust model for consistency.

The intrinsic GRB afterglow spectrum is believed to be emitted as synchrotron radiation from the interaction of the ultra-relativistic jet and the ISM. As a consequence, the intrinsic SED of the afterglow is expected to follow a power law with \(F_\nu = \nu^{-\beta}\). In some cases, a change in the spectral slope of \(\Delta \beta = 0.5\) between the optical and X-ray spectra is observed (Greiner et al. 2011; Zafar et al. 2011), known as the cooling break (Sari, Piran & Narayan 1998). The observed, dust extinguished spectrum can therefore be modelled as \(F_\nu^{\text{obs}} = F_\nu 10^{-\delta A_V}\), where \(A_V\) is the extinction due to dust absorption and scattering as a function of wavelength.

We used the spectral fitting package XSPEC v. 12.9 to fit the rest-frame optical to near-infrared SEDs of the GRB afterglows, assuming a single or broken power law together with the parametric dust model from Fitzpatrick & Massa (1990, hereafter F&M 90). In systems where a broken power law is preferred, a cooling break \(\nu_{\text{break}}\) is required between the intrinsic spectral optical (\(\beta_{\text{opt}}\)) and X-ray (\(\beta_X\)) slopes and were fitted such that the change in slope is fixed to \(\Delta \beta = 0.5\). The X-ray data are taken from the Swift archive facility (Evans et al. 2009) and for each case the spectrum in the 0.3–10 keV range is reduced around the SED mid-time in xselect. We then fix the total Galactic equivalent hydrogen column density, \(N_{\text{HGal}}\), for the X-ray spectra to the values from Willingale et al. (2013) but leave the rest-frame GRB host galaxy equivalent hydrogen column density, \(N_{\text{HISM}}\), as a free parameter. The F&M 90 dust model allows the individual extinction curves to be fit through a set of eight parameters and is defined as

\[
A_V = \frac{A_V}{R_V} (c_1 + c_2 x + c_3 D(x, x_0, \gamma) + c_4 F(x) + 1) ,
\]

where

\[
D(x, x_0, \gamma) = \frac{x^2}{(x^2 - x_0^2)^2 + x^2 \gamma^2}
\]

and

\[
F(x) = \begin{cases} 
0.539(x - 5.9)^2 + 0.056(x - 5.9)^3 & \text{for } x \geq 5.9 \\
0 & \text{for } x < 5.9
\end{cases}
\]

with \(x = \lambda^{-1}\) in units of \(\mu m^{-1}\). Basically, this dust model contains two components, one describing the linear UV part of the spectrum via the components \(c_1\) (intercept), \(c_2\) (slope), and the term \(c_4 F(x)\) describing the far-UV curvature. The second component is the Drude profile representing the 2175 Å extinction bump, controlled by the parameters \(c_3\) (bump strength), \(x_0\) (central wavelength), and \(\gamma\) (width of the bump). The last two parameters are the visual extinction, \(A_V\), and the total-to-selective reddening, \(R_V\). For the systems where the 2175 Å extinction bump is not detected, we fix the Drude components to \(\gamma = 1 \mu m^{-1}\) and \(x_0 = 4.6 \mu m^{-1}\) but leave \(c_3\) as a free parameter (even though Zafar et al. 2015, 2018a, found that fixing \(c_3 = 0\) reduces the \(\chi^2\)). We compute the 3σ upper limits on the bump strength instead, given by \(c_3\), to compare to the GRBs where the 2175 Å extinction bump is clearly detected.
Table 2. Gas-phase abundances of the GRB C I absorbers. References for the individual bursts and gas-phase abundances are given in the Appendix under the dedicated notes on each GRB.

<table>
<thead>
<tr>
<th>GRB</th>
<th>log N(H I) (cm$^{-2}$)</th>
<th>log N(Zn II) (cm$^{-2}$)</th>
<th>log N(Fe II) (cm$^{-2}$)</th>
<th>[Zn/H]</th>
<th>[Zn/Fe]</th>
<th>log N(Fe)$_{\text{dust}}$ (cm$^{-2}$)</th>
<th>log N(Si)$_{\text{dust}}$ (cm$^{-2}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>061121</td>
<td>$-0.05$ ± $0.06$</td>
<td>$-0.05$ ± $0.06$</td>
<td>$-0.05$ ± $0.06$</td>
<td>$0.51$</td>
<td>$0.07$</td>
<td>$15.55$ ± $0.06$</td>
<td>$-0.05$ ± $0.06$</td>
</tr>
<tr>
<td>070802</td>
<td>$0.25$ ± $0.20$</td>
<td>$0.25$ ± $0.20$</td>
<td>$0.25$ ± $0.20$</td>
<td>$0.45$</td>
<td>$0.05$</td>
<td>$16.36$ ± $0.41$</td>
<td>$-0.05$ ± $0.06$</td>
</tr>
<tr>
<td>080605</td>
<td>$-0.05$ ± $0.06$</td>
<td>$-0.05$ ± $0.06$</td>
<td>$-0.05$ ± $0.06$</td>
<td>$0.51$</td>
<td>$0.07$</td>
<td>$15.55$ ± $0.06$</td>
<td>$-0.05$ ± $0.06$</td>
</tr>
<tr>
<td>080607</td>
<td>$0.25$ ± $0.20$</td>
<td>$0.25$ ± $0.20$</td>
<td>$0.25$ ± $0.20$</td>
<td>$0.45$</td>
<td>$0.05$</td>
<td>$16.36$ ± $0.41$</td>
<td>$-0.05$ ± $0.06$</td>
</tr>
<tr>
<td>120119A</td>
<td>$0.25$ ± $0.20$</td>
<td>$0.25$ ± $0.20$</td>
<td>$0.25$ ± $0.20$</td>
<td>$0.45$</td>
<td>$0.05$</td>
<td>$16.36$ ± $0.41$</td>
<td>$-0.05$ ± $0.06$</td>
</tr>
<tr>
<td>120815A</td>
<td>$0.25$ ± $0.20$</td>
<td>$0.25$ ± $0.20$</td>
<td>$0.25$ ± $0.20$</td>
<td>$0.45$</td>
<td>$0.05$</td>
<td>$16.36$ ± $0.41$</td>
<td>$-0.05$ ± $0.06$</td>
</tr>
<tr>
<td>120815A</td>
<td>$0.25$ ± $0.20$</td>
<td>$0.25$ ± $0.20$</td>
<td>$0.25$ ± $0.20$</td>
<td>$0.45$</td>
<td>$0.05$</td>
<td>$16.36$ ± $0.41$</td>
<td>$-0.05$ ± $0.06$</td>
</tr>
<tr>
<td>121024A</td>
<td>$0.25$ ± $0.20$</td>
<td>$0.25$ ± $0.20$</td>
<td>$0.25$ ± $0.20$</td>
<td>$0.45$</td>
<td>$0.05$</td>
<td>$16.36$ ± $0.41$</td>
<td>$-0.05$ ± $0.06$</td>
</tr>
<tr>
<td>150403A</td>
<td>$0.25$ ± $0.20$</td>
<td>$0.25$ ± $0.20$</td>
<td>$0.25$ ± $0.20$</td>
<td>$0.45$</td>
<td>$0.05$</td>
<td>$16.36$ ± $0.41$</td>
<td>$-0.05$ ± $0.06$</td>
</tr>
<tr>
<td>180325A</td>
<td>$0.25$ ± $0.20$</td>
<td>$0.25$ ± $0.20$</td>
<td>$0.25$ ± $0.20$</td>
<td>$0.45$</td>
<td>$0.05$</td>
<td>$16.36$ ± $0.41$</td>
<td>$-0.05$ ± $0.06$</td>
</tr>
</tbody>
</table>

This value represents the lower limit on [O/H] measured from the equivalent width of O I λ 1355, following Prochaska et al. (2009).

Figure 1. Individual extinction curves of the GRB C I absorbers. Each curve has been colour-coded as a function of the total-to-selective extinction, $R_V$, violet representing a dust grain size distribution composed of small grains and red a dust composition with predominantly larger dust grain sizes. The 2175 Å dust extinction feature appears to be more prominent for larger values of $R_V$. For comparison, the average extinction curves observed in the MW, SMC, and LMC are also shown.

4 RESULTS

Overall, we find a broad range of distinct dust models characterizing the GRB C I absorbers. The individual best fit extinction curves are shown in Fig. 1. In Table 3 we list the set of best-fitting parameters for the F&M90 dust model for the GRBs 061121, 070802, 080605, 080607, and 150403A determined in this work, including previously published values for GRBs 120119A, 120815A, 121024A, and 180325A from Zafar et al. (2018a). We also provide $A_{\text{bump}} = \pi c / (2 \omega \lambda R_V) \times A_V$ (or the 3σ upper limit) for each of the GRB C I absorbers. The detection limit on the 2175 Å dust extinction feature is typically $A_{\text{bump}} < 0.2$ mag.

In our sample, four out of nine cases (∼45 per cent) show a prominent 2175 Å dust extinction feature (GRBs 070802, 080605, 080607, and 180325A). Six systems (∼65 per cent) show relatively steep extinction curves with $R_V = 2.4$–$3.0$ (GRBs 061121, 070802, 120119A, 120815A, 121024A, and 150403A), where the other three systems (∼35 per cent) show more ‘flat’ extinction curves with $R_V = 3.3$–$4.5$. We also measure a large variation in bump strength between the individual systems, in the range $A_{\text{bump}} = 0.2$–$2.5$ mag.

In general, we find that the amount of C I is connected to the dust properties inferred from the extinguished SED, such as $A_V$, $R_V$, and $A_{\text{bump}}$. To quantify this we show pairwise scatter plots and histograms of different dust tracers colour-coded as a function of $W(\lambda 1560)$ in Fig. 2. Due to the small sample size and relatively large error bars, the observed trends are only found to be correlated at 1–2σ significance. The connection of $A_V$ and $A_{\text{bump}}$ with C I is already well-established for quasar (Ledoux et al. 2015; Ma et al. 2018) and GRB (Zafar et al. 2012; Heintz et al. 2019) absorption systems, but here we include the additional information derived from the slope of the extinction, as inferred from $R_V$. The observed trend between $A_V$ and $W(\lambda 1560)$ indicates that larger amounts of cold gas is found in more dusty sightlines, where the additional trend with $R_V$ also suggests that the dust grain size distribution is on average larger in these sightlines as well. Contrary to this, we observe no relation between $A_V$, $R_V$ or the amount of C I to the typically used depletion-derived dust tracers such as the zinc-to-iron depletion level, $[\text{Zn/Fe}]$, and the dust-phase iron column density, $N(\text{Fe})_{\text{dust}}$. We caution that since it is not possible to infer the depletion of the two strongest C I absorbers (GRBs 080607 and 180325A), the apparent non-correlation between the amount of C I to $[\text{Zn/Fe}]$ and $N(\text{Fe})_{\text{dust}}$ might be biased. However, even without those systems we are still probing a large range of depletion values (i.e. $[\text{Zn/Fe}] = 0.5$–$2.0$), without seeing any trends with either $A_V$, $R_V$, or $A_{\text{bump}}$. Also, for the bursts where the depletion could be derived, the GRB showing the strongest extinction bump (GRB 070802) shows the lowest value of $[\text{Zn/Fe}]$ and $N(\text{Fe})_{\text{dust}}$. Finally, if we assume that e.g. GRB 080607 has solar metallicity (Prochaska et al. 2009), then the depletion of the system would be around $[\text{Zn/Fe}] \sim 1.1$ following the relation from Ledoux, Petitjean & Srianand (2003), which would still not produce any trend with $A_V$, $R_V$, $A_{\text{bump}}$, or $W(\lambda 1560)$. We therefore argue that, even based on the small sample size, the amount of C I and SED-derived dust properties is unlikely to be connected to the depletion-derived dust content for these particular absorbers. In the following section we will attempt to provide a resolution to this apparent discrepancy.
Table 3. The best-fitting F&M 90 extinction curve parameters for the X-ray to the optical/near-infrared afterglow SEDs of the GRB C I absorbers.

<table>
<thead>
<tr>
<th>GRB</th>
<th>$c_1$</th>
<th>$c_2$</th>
<th>$c_3$</th>
<th>$c_4$</th>
<th>$\gamma$</th>
<th>$\chi^2_0$</th>
<th>$R_V$</th>
<th>$A_V$ (mag)</th>
<th>$A_{\text{bump}}$ (mag)</th>
</tr>
</thead>
<tbody>
<tr>
<td>061121</td>
<td>$-4.63 \pm 0.13$</td>
<td>$2.11 \pm 0.11$</td>
<td>$&lt;0.46$</td>
<td>$0.34 \pm 0.12$</td>
<td>$1.00$</td>
<td>$4.60$</td>
<td>$2.88^{+0.23}_{-0.27}$</td>
<td>$0.46^{+0.11}_{-0.15}$</td>
<td>$&lt;0.16$</td>
</tr>
<tr>
<td>070802</td>
<td>$0.03 \pm 0.33$</td>
<td>$0.88 \pm 0.17$</td>
<td>$1.56 \pm 0.27$</td>
<td>$0.45 \pm 0.09$</td>
<td>$0.99 \pm 0.01$</td>
<td>$4.63 \pm 0.01$</td>
<td>$2.72^{+0.41}_{-0.54}$</td>
<td>$1.20^{+0.15}_{-0.15}$</td>
<td>$1.09 \pm 0.34$</td>
</tr>
<tr>
<td>080605</td>
<td>$-6.12 \pm 0.84$</td>
<td>$2.66 \pm 0.39$</td>
<td>$0.46 \pm 0.11$</td>
<td>$1.84 \pm 0.50$</td>
<td>$0.60 \pm 0.08$</td>
<td>$4.53 \pm 0.01$</td>
<td>$3.27^{+0.88}_{-0.92}$</td>
<td>$0.48^{+0.13}_{-0.12}$</td>
<td>$0.18 \pm 0.08$</td>
</tr>
<tr>
<td>080607</td>
<td>$1.14 \pm 0.34$</td>
<td>$0.36 \pm 0.11$</td>
<td>$3.05 \pm 0.72$</td>
<td>$0.17 \pm 0.05$</td>
<td>$1.22 \pm 0.05$</td>
<td>$4.51 \pm 0.02$</td>
<td>$4.14^{+1.05}_{-0.90}$</td>
<td>$2.58^{+0.42}_{-0.45}$</td>
<td>$2.45 \pm 0.97$</td>
</tr>
<tr>
<td>120119A</td>
<td>$-4.13 \pm 0.08$</td>
<td>$2.09 \pm 0.07$</td>
<td>$&lt;0.43$</td>
<td>$0.22 \pm 0.10$</td>
<td>$1.00$</td>
<td>$4.60$</td>
<td>$2.91^{+0.24}_{-0.18}$</td>
<td>$1.02^{+0.11}_{-0.11}$</td>
<td>$&lt;0.26$</td>
</tr>
<tr>
<td>120815A</td>
<td>$-4.77 \pm 0.08$</td>
<td>$2.14 \pm 0.07$</td>
<td>$&lt;0.32$</td>
<td>$0.82 \pm 0.08$</td>
<td>$1.00$</td>
<td>$4.60$</td>
<td>$2.39^{+0.09}_{-0.09}$</td>
<td>$0.19^{+0.04}_{-0.04}$</td>
<td>$&lt;0.05$</td>
</tr>
<tr>
<td>121024A</td>
<td>$-4.23 \pm 0.06$</td>
<td>$2.20 \pm 0.08$</td>
<td>$&lt;0.35$</td>
<td>$0.57 \pm 0.05$</td>
<td>$1.00$</td>
<td>$4.60$</td>
<td>$2.81^{+0.20}_{-0.16}$</td>
<td>$0.26^{+0.07}_{-0.07}$</td>
<td>$&lt;0.06$</td>
</tr>
<tr>
<td>150403A</td>
<td>$-4.83 \pm 0.08$</td>
<td>$2.23 \pm 0.05$</td>
<td>$&lt;0.35$</td>
<td>$0.59 \pm 0.02$</td>
<td>$1.00$</td>
<td>$4.60$</td>
<td>$&lt;2.81$</td>
<td>$&lt;0.13$</td>
<td>$&lt;0.03$</td>
</tr>
<tr>
<td>180325A</td>
<td>$-1.95 \pm 0.32$</td>
<td>$1.28 \pm 0.17$</td>
<td>$2.92 \pm 0.19$</td>
<td>$0.52 \pm 0.19$</td>
<td>$1.16 \pm 0.06$</td>
<td>$4.54 \pm 0.03$</td>
<td>$4.58^{+0.37}_{-0.39}$</td>
<td>$1.58^{+0.10}_{-0.12}$</td>
<td>$1.36 \pm 0.19$</td>
</tr>
</tbody>
</table>

Note. The best-fitting curve coefficients are from aZafar et al. (2018a), bZafar et al. (2018c) or derived in this work. The strength of the 2175 Å extinction bump is calculated as $A_{\text{bump}} = \pi c_1/(2\gamma R_V) \times A_V$. The reduced $\chi^2$, the number of degrees of freedom (dof), the null hypothesis probability (NHP) for the best fit, and the F-test probability is given in the Appendix for each individual GRB.

Figure 2. Pairwise scatter plots showing a comparison of different dust tracers, colour-coded as a function of the rest-frame equivalent width of C I $\lambda$ 1560. The SED-derived dust properties, i.e. $A_V$, $R_V$, and $A_{\text{bump}}$ are compared to the commonly used depletion-derived dust tracers, the zinc-to-iron depletion, $[\text{Zn/Fe}]$, and the dust-phase iron column density, $N(\text{Fe})_{\text{dust}}$. Positive trends with C I are mainly observed for the SED-derived dust properties but also tentatively for the metallicity, $[\text{Zn/H}]$ (see main text). For GRB 080607 we show the estimated lower limit on the $[\text{O/H}]$-derived metallicity instead of $[\text{Zn/H}]$, as also listed in Table 2.
5 DISCUSSION

5.1 The presence of carbon-rich dust in molecular clouds

The previously observed trend between $\rho_{\text{V}}$ and the amount of $\text{C}_1$ indicates that whenever the GRB sightline intersects a molecular cloud, the amount of cold gas ($\text{C}_1$ in this case) is connected to the amount of dust derived from the extinction. Including the additional trend with $R_{\gamma}$ observed in this work, it is clear that the average grain size distribution of the dust particles in the molecular cloud are also related to these properties. This would suggest that the $\text{C}_1$ systems are dust-rich with a composition dominated by carbonaceous material (as already speculated in Paper I), that the dust grain size distribution mainly consists of large dust grains, and that this carbon-rich dust dominates the shape of the extinguished SED. Here we argue for a simple scenario to explain these relations: The amount of $\text{C}_1$ simply reflects how deep into the intervening molecular cloud the GRB sightline probes, where we expect larger dust columns, dust grains and higher $\text{C}_1$ column densities to be present closer to the centre of the cloud (Bolatto, Wolfire & Leroy 2013). Below we will outline some of the key points that support this scenario.

First, we argue that when cold and molecular gas is detected in GRB sightlines (i.e. from absorption features of $\text{C}_1$ and/or $\text{H}_2$) it is not related to the molecular gas associated with the GRB progenitor star. It has been found that the intense $\gamma$-ray flash, the afterglow emission or UV photons from the natal HII region of the GRB will not be related to the molecular gas associated with the GRB progenitor star. It has been found that the intense $\gamma$-ray flash, the afterglow emission or UV photons from the natal HII region of the GRB will not be related to the molecular gas associated with the GRB progenitor star but is likely only connected to the molecular gas. We will argue that the anticorrelation with $R_{\gamma}$ is likely only connected to the non-carbonaceous dust grains. In this scenario, $R_{\gamma}$ is likely only connected to the non-carbonaceous dust grains. If this scenario is true, we predict that $\rho_{\text{V}}$ is only correlated to the non-carbonaceous dust tracers [such as $N(\text{Fe}^\text{dust})$; Vladilo et al. 2006; De Cia et al. 2013] if the GRB sightline does not intersect a molecular cloud. If instead, a molecular cloud consisting of significant amounts of carbon (in the form of $\text{C}_1$ or CO) intervenes the GRB and quasar sightline, the carbon-rich dust will significantly change the shape of the extinction curve, such that $N(\text{Fe}^\text{dust})$ or the depletion-derived $R_{\gamma}$ is no longer a good representation of the integrated dust composition but only contributes mildly to the overall shape of the extinguished SED. This could also explain why Bolmer et al. (2018) found that there is more carbon-rich dust in GRB hosts at $z > 4$, simply due to the smaller amounts of dust grains formed from grain growth in the ISM at this epoch.

Finally, we note that Ramírez-Tamus et al. (2018) found a correlation between the strength of diffuse interstellar bands (DIBs; e.g. Cami & Cox 2014) with the amount of dust towards the star-forming region M17, but an anticorrelation with $R_{\gamma}$. The strongest DIBs are, however, believed to originate in the diffuse ISM, such that the anticorrelation with $R_{\gamma}$ is only likely connected to the ‘warm’ gas-phase of the ISM and not to the molecular gas. We will discuss this further in Section 5.3.

5.2 Dust composition of molecular clouds at high-$z$

Following the arguments laid out above, it is now possible to constrain the dust composition and grain size distribution of these intervening, high-$z$ molecular clouds. First, we note from Fig. 3 that the variation in the grain-size distribution observed in this work support a model where small dust grains condense into large grains in increasingly dense environments (Draine 1990; Weingartner & Draine 2001). The relatively steep extinction curves observed towards GRBs 061121, 070802, 120119A, 120815A, 121024A, and 150403A indicate a grain size distribution composed mainly of small ($\lesssim 0.25 \mu m$) silicate grains in the ISM (Mishra & Li 2017). The extinction curves observed toward GRBs 080605, 080607, and 180325A, however, require large average grain sizes ($\approx 0.4$–$0.5 \mu m$) and a significant contribution from small carbonaceous dust grains

and Large Magellanic Clouds (SMC and LMC), the Milky Way (MW; Gordon et al. 2003; Fitzpatrick & Massa 2007) or in average high-redshift GRB absorbers (excluding bursts with $\text{C}_1$ and/or $\text{H}_2$ detections; Zafar et al. 2018a,b). Since $\rho_{\text{V}}$ and $R_{\gamma}$ appear to be correlated in our sample of GRB $\text{C}_1$ absorbers, it is a reasonable assumption that they are regulated by the same physical processes and thus originates from the same molecular cloud. This further supports that the SED-derived dust properties in GRB $\text{C}_1$ absorbers characterize the dust composition and grain size distribution of the dust in the molecular gas-phase and not in ISM of the GRB host galaxies.

The amount of $\text{C}_1$ is then expected to correlate with the SED-derived dust extinction, but not necessarily with other dust tracers such as the sun dust depletion, [Zn/Fe], and the dust-phase iron column density, $N(\text{Fe}^\text{dust})$ (as observed, see Fig. 2). Since dust can form if there are sufficient dense and cold gas available, non-carbonaceous dust grains will specifically form via grain growth in the ISM if there is a large reservoir of metals available as well (mostly O, Fe, Si, and Mg; Draine 2003), whereas carbon-rich dust will form if there is instead sufficient carbon available. Based on this, we therefore argue that in the ISM, the amount of dust is well-represented by either the derived depletion level, e.g. [Zn/Fe] (De Cia 2018) and/or the dust-phase iron abundance, since these quantities trace non-carbonaceous dust. If this scenario is true, we predict that $\rho_{\gamma}$ is only correlated to the non-carbonaceous dust tracers [such as $N(\text{Fe}^\text{dust})$; Vladilo et al. 2006; De Cia et al. 2013] if the GRB and quasar sightline does not intersect a molecular cloud. If instead, a molecular cloud consisting of significant amounts of carbon (in the form of $\text{C}_1$ or CO) intervenes the GRB and quasar sightline, the carbon-rich dust will significantly change the shape of the extinction curve, such that $N(\text{Fe}^\text{dust})$ or the depletion-derived $\rho_{\gamma}$ is no longer a good representation of the integrated dust composition but only contributes mildly to the overall shape of the extinguished SED. This could also explain why Bolmer et al. (2018) found that there is more carbon-rich dust in GRB hosts at $z > 4$, simply due to the smaller amounts of dust grains formed from grain growth in the ISM at this epoch.

Finally, we note that Ramírez-Tamus et al. (2018) found a correlation between the strength of diffuse interstellar bands (DIBs; e.g. Cami & Cox 2014) with the amount of dust towards the star-forming region M17, but an anticorrelation with $R_{\gamma}$. The strongest DIBs are, however, believed to originate in the diffuse ISM, such that the anticorrelation with $R_{\gamma}$ is only likely connected to the ‘warm’ gas-phase of the ISM and not to the molecular gas. We will discuss this further in Section 5.3.
Based on a sample of quasar absorbers selected solely on the basis of the 2175 Å dust extinction feature, Ma et al. (2018) found that all such systems are associated with strong absorption features of the 2175 Å dust extinction feature, Ma et al. (2018) found that the carriers producing the 2175 Å dust extinction feature is also not located in the ‘warm’ neutral gas-phase of the ISM.

If the 2175 Å dust extinction feature is instead connected to the molecular gas-phase of the ISM, we would expect from the scenario outlined in the previous sections that the detection probability and strength of this feature is better correlated with the SED-derived dust properties. In GRB and quasar sightlines, the detection and strength of the 2175 Å dust extinction feature has previously been found to be connected to dusty sightlines (Ledoux et al. 2015; Corre et al. 2018; Ma et al. 2018) and the amount of C I in the absorbing systems (Zafar et al. 2012; Ledoux et al. 2015; Ma et al. 2018; Heintze et al. 2019). In Fig. 5, we explore the connection between the characteristic bump and the grain size distribution, represented by $R_V$ (Draine 2003). We find that the carriers producing the 2175 Å dust extinction feature in GRB hosts appears to be more prominent in systems with large amounts of C I and a grain size distribution composed mainly of large dust grains. The quasar C I absorber population appears to also be consistent with this, though only limited information is available on the slope of the extinction, $R_V$ (Ledoux et al. 2015). If $R_V$ and C I is indeed connected to large dust grains and cold neutral gas observed deep into molecular clouds, the 2175 Å dust extinction feature must by association be produced in the same molecular cloud.

Finally, Cortzen et al. (2019) recently found that PAHs are connected to the molecular gas-phase in star-forming galaxies. If PAHs then contribute to or are the sole carrier of the 2175 Å dust extinction feature (as argued by Li & Draine 2001; Draine & Li 2007), this would further support the scenario where the bump is produced by carriers located in molecular clouds. In addition, since the bump strength is not connected to the presence of DIBs (Xiang, Li & Zhong 2011), this excludes the scenario where the carriers of the 2175 Å dust extinction feature is located in the ‘warm’ neutral ISM. This would also explain why GRB 070802, showing a strong 2175 Å extinction bump, stands out as a clear outlier in the relation between $A_V$ and N(Fe)$_{dust}$ (De Cia et al. 2013), simply because a significant dust component from the carbon-rich dust in the molecular cloud affects the shape of the SED but not the gas-phase depletions.
we found no connection between the amount of C I to the dust depletion, [Zn/Fe], or the dust-phase iron abundance, \( N(\text{Fe})_{\text{dust}} \), but note a tentative correlation with the gas-phase metallicity, [Zn/H]. We, therefore, further advocate against using the depletion-derived dust content as a tracer of the total line-of-sight extinction (as also previously cautioned; Savaglio & Fall 2004; Wiseman et al. 2017), at least when the individual dust components cannot be securely identified.

We discussed a scenario where the main dust component causing the observed line-of-sight extinction in the GRB C I absorbers is located in the intervening, C I-bearing molecular cloud. We argued that such a scenario could explain the connection of C I with only the SED-derived dust properties, and reconcile the apparent discrepancy between the extinction and depletion-derived dust properties. Here, the main component causing the observed extinction is carbon-rich dust in the molecular cloud which is not sensitive to the amount of depleted, non-carbonaceous metals in the dust-phase of the ISM. Consistently, we found that the detection and strength of the 2175 Å dust extinction feature is also not linked to any of the non-carbonaceous dust indicators, but better correlated with \( A_V \), \( R_V \), and the amount of C I. Moreover, the characteristic dust bump appears to be possibly linked to carbonaceous dust grains and the molecular gas-phase of the ISM, supporting PAHs as potential carriers of the 2175 Å dust extinction feature. We also note that Mishra & Li (2017) did not find any correlation between the extinction parameters and silicon in the dust-phase, but did find that the 2175 Å dust bump scales with the amount of carbon in the dust-phase in Galactic sightlines. This is consistent with the above interpretation of the GRB absorbers showing significant C I abundances.

The evidence for the validity of such a scenario in high-redshift C I-bearing absorbers as presented here, is still only based on tentative correlations and should be verified if more GRB C I systems, especially with the 2175 Å dust extinction feature, are observed. Modelling the individual contributions from a large dust component located in the molecular gas-phase of the ISM and a secondary component in the 'warm' neutral medium would also be beneficial to further understand the observed dust content of high-redshift star-forming galaxies.
ACKNOWLEDGEMENTS

We would like to thank the referee for providing a clear, constructive, and concise report that greatly improved the interpretations presented here. We would also like to thank Karl Gordon for an enlightening discussion in the early stages of this work. KEH and PJ acknowledge support by a Project Grant (162948-051) from The Icelandic Research Fund. The Cosmic Dawn Center is funded by the Danish National Research Foundation (DNRF).

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APPENDIX A: NOTES ON INDIVIDUAL GRBs

GRB 061121

This GRB was first detected by Swift and reported by Page et al. (2006). For this case we derived the extinction curve parameters listed in Table 3 and the best fit model and $A_V$ are shown in Fig. A1. The spectra examined in this work are obtained with the Keck/LRIS spectrograph and published by Fynbo et al. (2009). The gas-phase abundances listed in Table 2 are from Zafar & Möller (2019). In addition to the parametric dust model we also obtain the best fit for the equivalent neutral hydrogen column density of $N_{HI} < 0.94 \times 10^{22}$ cm$^{-2}$, the optical slope, $\beta_{opt} = 0.41 \pm 0.10$, the X-ray slope, $\beta_X = 0.91 \pm 0.11$, and the break frequency, $\nu_{\text{break}} = 16.70 \pm 0.15$ Hz. The SED fits well with a broken power-law (with an $F$-test probability of < 0.01) and the resulting reduced $\chi^2$ together with the number of degrees of freedom is $\chi^2/(\text{dof}) = 1657/6161$, with a null hypothesis probability of 100 per cent for the best fit model. These results are consistent with the best fit models derived by Schady et al. (2010) and Covino et al. (2013), which are based solely on photometry.

GRB 070802

This GRB was first detected by Swift and reported by Barthelmy et al. (2007). For this case we derived the extinction curve parameters listed in Table 3 and the best-fitting model and $A_V$ are shown in Fig. A2. The spectrum examined in this work is from the VLT/FORS2 spectrograph and published by Elíasdóttir et al. (2009) and Fynbo et al. (2009). In addition to the parametric dust model we also obtain the best fit for the equivalent neutral hydrogen column density of $N_{HI} < 2.91 \times 10^{22}$ cm$^{-2}$. The SED fits well with a single power law (with an $F$-test probability of 1.00) with a slope of $\beta_{opt,X} = 0.88 \pm 0.06$ and the resulting reduced $\chi^2$ together with the number of degrees of freedom is $\chi^2/(\text{dof}) = 750/1382$, with a null hypothesis probability of 100 per cent for the best fit model. This GRB shows a clear presence of the 2175 Å extinction bump (as first reported by Krühler et al. 2008; Elíasdóttir et al. 2009) and Zafar et al. (2011) have fitted the parametric dust model prescribed by Fitzpatrick & Massa (2007) to the same optical afterglow examined here. Our results are consistent with their best-fitting model, but we will use the results from fitting the SED with the F&M 90 parametrization in this work for consistency.

GRB 080605

This GRB was first detected by Swift and reported by Sbarufatti et al. (2008). For this case we derived the extinction curve parameters listed in Table 3 and the best-fitting model and $A_V$ are shown in Fig. A3. The spectrum examined in this work is obtained with the VLT/FORS2 spectrograph and published by Fynbo et al.
This GRB was first detected by \textit{Swift} and reported by Beardmore et al. (2012). The data presented here were obtained with the VLT/X-shooter spectrograph, published by Selsing et al. (2019) and have been examined by Japelj et al. (2015) and Zafar et al. (2018a). We adopt the extinction curve parameters from Zafar et al. (2018a, see Table 3) since they fit for the full F&M 90 parametrization (whereas Japelj et al. 2015, only fit for $A_V$ assuming either an MW, SMC or LMC-like extinction curve). While both found a significant visual extinction, Japelj et al. (2015) claimed a detection of the 2175 Å extinction bump, although the best-fitting LMC extinction curve overpredicts the strength of the bump. Modelling the full F&M 90 parametric extinction law does not show a significant 2175 Å bump, however (see Zafar et al. 2018a), and we are thus only able to provide upper limits on $A_{\text{bump}}$. We adopt the column densities of H\textsc{i} and the low-ionization lines listed in Table 2 from Wiseman et al. (2017).
a null hypothesis probability of 100 per cent for the best-fitting model.

Figure A5. Same plot connotation as in Fig. A1 but for GRB 150403A observed with the VLT/X-shooter. The blue, green, and red colours correspond to the UVB, VIS, and NIR arm spectra, respectively.

This GRB was first detected by Swift and reported by Troja et al. (2018). The data presented here were obtained with the VLT/X-shooter spectrograph and published and examined by Zafar et al. (2018c). We adopt the extinction curve parameters listed in Table 3 and the column densities of H\textsc{i} and the low-ionization lines listed in Table 2 from their work. This GRB exhibit a clear presence of the 2175 Å extinction bump, first noted in the low-resolution NOT/ALFOSC spectra by Heintz, Fynbo & Malesani (2018b) obtained only 11 min post-burst. Subsequently, D’Avanzo et al. (2018) followed up this GRB with the VLT/X-shooter, confirming the detection of the 2175 Å extinction bump and detected several absorption lines and the nebular emission lines [O\textsc{ii}], [O\textsc{iii}], and H\textalpha. Zafar et al. (2018c) found that the four different epochs of the GRB afterglow show consistent dust properties. We will use the best fit extinction curve parameters from the first epoch VLT/X-shooter spectrum in this work.