Fig. S1. FTIR spectra of Cr-MIL-101-NO₂ (top) and Cr-MIL-101-NH₂ (bottom). The characteristic signals are marked with an asterisk for clarity.

Fig. S2. FTIR spectra of Cr-MIL-101-NO₂ (top) and Cr-MIL-101-NH₂ (bottom). The characteristic signals are marked with an asterisk for clarity.
Fig. S3. $^1$H-NMR spectra of the linker molecules. Left: after 10 minutes nitration time, the signal at 7.3 ppm (marked by an asterisk) belongs to unreacted terephthalic acid. Integration of the signals show a molar ratio terephthalic acid : nitrotetraphthalic acid = 1:6. Right: after five hours reaction time. There is no signal of the terephthalic acid left. Expanding the reaction time up to 48 h no multiple nitration products were observed.

Fig. S4. $^1$H-NMR spectrum of the linker after the reduction of Cr-MIL-101-NO$_2$ to Cr-MIL-101-NO$_2$. Only aminoterephthalic acid is observed.
**Fig. S5** IR spectra of Cr-MIL-101-NH$_2$ (bottom) and the urea form Cr-MIL-101-UR2 (top). The characteristic signals are marked with asterisks for clarity.

**Fig. S6.** $^{13}$C-NMR spectrum of the linker from the product (Cr-MIL-101-UR2) which was obtained from the reaction of Cr-MIL-101-NH$_2$ with ethyl isocyanate. The spectrum was recorded in NaOD/D$_2$O (20%). All signals can be clearly assigned.
Fig. S7. TGA data of the investigated compounds Cr-MIL-101-NO$_2$(1), Cr-MIL-101-NO$_2$(2), Cr-MIL-101-NH$_2$ and Cr-MIL-101-UR2.
Fig. S8. $^1$H-NMR spectrum of the linker of Cr-MIL-101-UR measured in NaOD/D$_2$O (20%). The assignments of the signals can be found in Fig. 5. The integrals can be clearly assigned to the corresponding protons.