Electronic Supplementary Material (ESI) for Dalton Transactions.

Supplementary Table Captions STable 1. Atomic coordinates (x,y,z), isotropic displacement parameters and site occupancy factors and their estimated standard deviations found with the Rietveld refinement results from bulk samples of BaxCr5Se8

STable 2. Main distances extracted from Rietveld refinement on bulk samples of BaxCr5Se8

Supplementary Figure Caption:

SFigure 1. Temperature dependence of the magnetic susceptibility for BaxCr5Se8 measured under a magnetic field of 0.1 T for x = 0.5, 0.55 and 0.6. An antiferromagnetic transition is observed at 58 K, where the derivative of the curve is maximum. Only the 0.6 content sample shows a ferrimagnetic transition, with a transition temperature tending to the one of Ba1-pCr2Se4-p.

STable 1. Atomic coordinates (x,y,z), isotropic displacement parameters and site occupancy factors and their estimated standard deviations found with the Rietveld refinement results from bulk samples of $Ba_xCr_5Se_8$

	x = 0.5					x = 0.51					x = 0.52					x = 0.55				
Atoms	s.o.f	x	y	z	U_{iso}	s.o.f	x	y	z	U_{iso}	s.o.f	x	y	z	U_{iso}	s.o.f	x	y	z	U_{iso}
Ba(1)	0.874(7)	0	0	0	0.020(4)	0.906(7)	0	0	0	0.019(3)	0.904(7)	0	0	0	0.031(3)	0.861(8)	0	0	0	0.043(3)
Ba(2)	0.126(7)	0	0.5	0	0.025(4)	0.114(7)	0	0.5	0	0.019(3)	0.136(7)	0	0.5	0	0.031(3)	0.239(8)	0	0.5	0	0.043(3)
Cr(1)	1	0.996(2)	0.749(3)	0.510(2)	0.004(4)	1	0.999(2)	0.752(3)	0.506(3)	-0.001(4)	1	1.000(2)	0.746(3)	0.506(2)	0.004(3)	1	0.998(3)	0.745(3)	0.509(2)	0.006(3)
Cr(2)	1	0.5894(18)	0.397(5)	0.1543(17)	0.011(2)	1	0.5903(19)	0.399(5)	0.1559(17)	0.0059(17)	1	0.5909(19)	0.399(5)	0.1621(16)	0.0103(16)	1	0.590(2)	0.397(4)	0.1595(16)	0.0098(15)
Cr(3)	1	0.685(2)	-0.082(6)	0.5285(19)	0.014(3)	1	0.6862(19)	-0.080(5)	0.5286(17)	0.008(3)	1	0.688(2)	-0.076(6)	0.5210(16)	0.012(2)	1	0.686(2)	-0.083(4)	0.5230(17)	0.013(2)
Cr(4)	1	0.407(2)	0.096(5)	-0.1761(18)	0.011(2)	1	0.411(2)	0.101(5)	-0.1705(18)	0.0059(17)	1	0.411(2)	0.095(5)	-0.1692(16)	0.0103(16)	1	0.407(2)	0.091(4)	-0.1705(17)	0.0098(15)
Cr(5)	1	0.686(2)	0.419(6)	0.515(2)	0.014(3)	1	0.688(2)	0.423(5)	0.5129(18)	0.008(3)	1	0.684(2)	0.417(6)	0.5200(16)	0.012(2)	1	0.683(2)	0.412(4)	0.5197(17)	0.013(2)
Se(1)	1	0.8527(15)	0.969(4)	0.3330(13)	0.006(2)	1	0.8511(15)	0.966(3)	0.3388(13)	0.0026(18)	1	0.8526(14)	0.965(4)	0.3352(12)	0.0037(16)	1	0.8545(15)	0.968(3)	0.3341(13)	0.0024(16)
Se(2)	1	0.4832(14)	0.375(4)	0.6548(13)	0.0047(19)	1	0.4835(13)	0.375(3)	0.6576(13)	-0.0006(17)	1	0.4770(12)	0.372(4)	0.6627(11)	0.0043(16)	1	0.4777(13)	0.371(3)	0.6631(12)	0.0029(16)
Se(3)	1	0.6639(13)	0.169(4)	-0.0009(14)	0.0091(17)	1	0.6669(14)	0.166(4)	0.0045(15)	0.0050(15)	1	0.6639(13)	0.163(4)	0.0113(12)	0.0046(14)	1	0.6643(13)	0.160(3)	0.0124(12)	0.0035(13)
Se(4)	1	0.8290(15)	0.711(3)	0.6797(14)	0.0072(19)	1	0.8319(14)	0.708(3)	0.6812(13)	0.0032(18)	1	0.8309(13)	0.708(3)	0.6780(12)	0.0048(16)	1	0.8319(14)	0.712(3)	0.6768(13)	0.0044(15)
Se(5)	1	0.8532(15)	0.464(4)	0.3467(13)	0.006(2)	1	0.8560(15)	0.469(3)	0.3392(12)	0.0026(18)	1	0.8523(14)	0.467(4)	0.3419(11)	0.0037(16)	1	0.8502(14)	0.470(3)	0.3433(12)	0.0024(16)
Se(6)	1	0.4771(14)	-0.129(4)	0.6584(12)	0.0041(19)	1	0.4785(13)	-0.130(3)	0.6572(12)	-0.0006(17)	1	0.4823(12)	-0.132(4)	0.6494(11)	0.0043(16)	1	0.4824(13)	-0.129(3)	0.6508(11)	0.0029(16)
Se(7)	1	0.3318(13)	0.327(4)	-0.0111(12)	0.0091(17)	1	0.3327(14)	0.326(4)	-0.0046(14)	0.0050(15)	1	0.3315(13)	0.326(4)	0.0003(11)	0.0046(14)	1	0.3330(14)	0.324(3)	0.0023(12)	0.0035(13)
Se(8)	1	0.8320(15)	0.202(3)	0.6835(13)	0.0075(19)	1	0.8290(14)	0.204(3)	0.6825(13)	0.0032(18)	1	0.8294(13)	0.205(3)	0.6849(12)	0.0048(16)	1	0.8277(14)	0.206(3)	0.6852(13)	0.0044(15)

* U_{iso} fixed for Ba(1)-Ba(2), Cr(2)-Cr(4), Cr(3)-Cr(5), Se(1)-Se(5), Se(2)-Se(6), Se(3)-Se(7) and Se(4)-Se(8) as explicited in the main paper.

STable 2. Main distances extracted from Rietveld refinement on bulk samples of $Ba_xCr_5Se_8$

	at 1	at	x = 0.5 [12]	x = 0.51 [This work]	x = 0.52 [This work]	x = 0.55 [This work]
tunneled Ba-Se	Ba(1)	Se(4)	3.402(15)	3.402(14)	3.422(15)	3.414(13)
		Se(8)	3.359(16)	3.387(15)	3.374(15)	3.381(14)
	Ba(2)	Se(4)	3.429(16)	3.399(15)	3.425(15)	3.442(14)
		Se(8)	3.409(15)	3.411(14)	3.391(14)	3.388(13)
lowest and largest In-plane and in-chain Cr-Se	Cr(1)	Se(1)		2.45(3)		
		Se(4)	2.43(3)		2.46(3)	2.41(3)
		Se(8)	2.64(3)	2.60(3)	2.62(3)	2.66(3)
	Cr(2)	Se(3)		2.47(4)	2.47(4)	2.46(3)
		Se(7)	2.46(4)			
		Se(6)	2.64(4)	2.65(3)	2.66(3)	2.67(3)
	Cr(3)	Se(4)	2.40(4)			2.41(3)
		Se(8)		2.44(3)	2.46(4)	
		Se(2)	2.66(4)	2.70(3)	2.68(4)	2.64(3)
lowest In-plane and in-chain Cr-Cr	Cr(1)	Cr(1)	3.60(3)	3.56(3)	3.55(3)	3.54(3)
		Cr(3)	3.44(4)	3.47(4)	3.48(4)	3.45(4)
		Cr(5)	3.42(4)	3.44(3)	3.49(3)	3.49(3)
	Cr(2)	Cr(2)	3.36(3)	3.38(3)	3.46(3)	3.44(3)

		Cr(4)	3.53(3)	3.49(3)	3.55(6)	3.54(3)
	Cr(3)	Cr(3)	3.83(4)	3.84(3)	3.86(4)	3.87(4)
		Cr(5)	3.59(6)	3.59(5)	3.55(6)	3.57(5)
	Cr(4)	Cr(4)	3.61(3)	3.55(3)	3.49(3)	3.51(3)
bridging	Cr(2)	Cr(5)	3.12(2)	3.09(2)	3.10(2)	3.12(2)
connection Cr-Cr	Cr(3)	Cr(4)	3.05(2)	3.10(2)	3.045(19)	3.05(2)