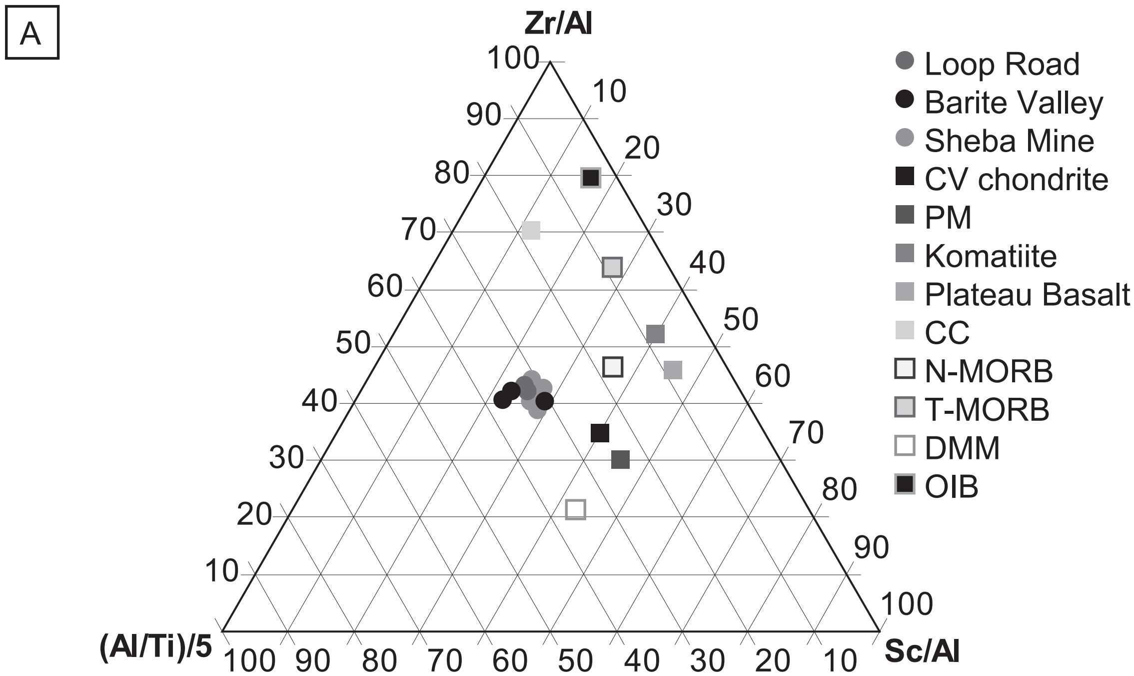

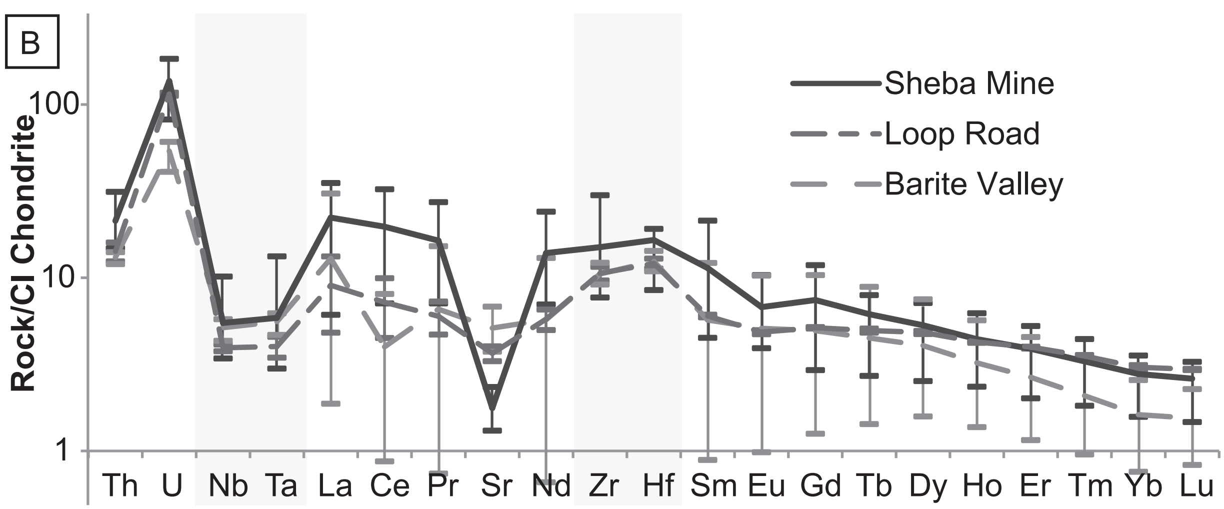

Figure DR1: Selection of the elements used in the model was based largely on prior work by the authors on the fallout sequence of spherules from the S3 layer (Krull-Davatzes et al., 2006) and from the diagenesis of the S3 layer at 8 locations (Krull-Davatzes et al., 2012). Full geochemical results are published in these papers, including data from underlying and overlying units. The three locations where a full fallout deposit was identified were used in this study, and these include the Barite Syncline, Sheba Mine, and Loop Road sections. Because some amount of variation occurs from base to top of a fallout layer, bulk analysis from the full thickness of each of these units were used. All have been deposited in different diagenetic environments with different over- and under-lying lithologies, so therefore we are able to determine the suite of unaltered elements for study. In addition, because all of these layers are fall deposits, we can eliminate transport fractionation (Krull-Davatzes et al., 2012). We deliberately selected a full range of elements that go from highly incompatible (Nb, Ta) to highly compatible (Cr, Ir) but immobile during diagenesis and metamorphism, as identified in Krull-Davatzes et al. (2012) and references therein. A) Ternary diagram of ratios of the elements Al, Ti, Sc, Zr, which tightly cluster for the S3 samples but are distinct from other endmember compositions. B) High field strength (HFSE) and rare earth elements (REE) from the three S3 sections display tight clustering and near identical averages for Nb, Ta, Zr and Hf (highlighted) but clear differences in the REEs due to carbonate and phosphate diagenesis, particularly in the Sheba Mine section (Krull-Davatzes et al., 2012). Authigenic U (Ua = U–Th/3) is only 0.3 ppm in the Barite Valley section, in contrast to 0.8 ppm and 0.9 ppm in the Loop Road and Sheba Mine sections, respectively. This is interpreted to be due to the differences in the depth of water of deposition and local redox conditions (Krull-Davatzes et al., 2012 and references therein) and we therefore removed these from our model.

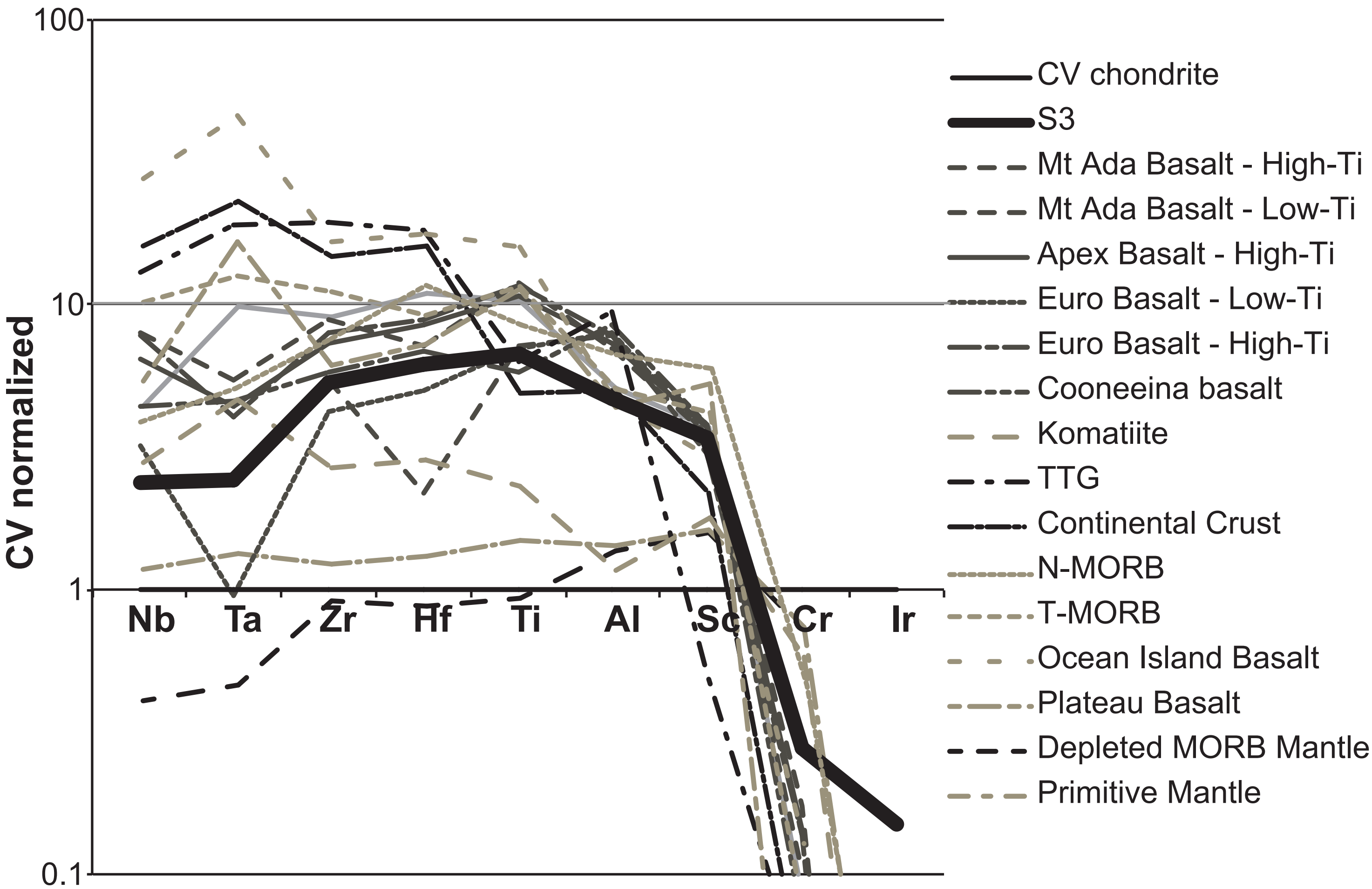

Figure DR2: Spidergrams of elements used within the mixing model of all crust and mantle endmembers, normalized to CV chondrite. Data for endmembers from Arevalo & Mcdonough, 2010; Ash et al., 2005; Kemp & Hawkesworth, 2003; Kerr, 2003; Klein, 2003; Lodders & Fegley, 1998; Maier et al., 2005; Palme & Jones, 2003; Palme & O'Neill, 2003; Rudnick & Gao, 2003; Salters & Stracke, 2004; Smithies et al., 2005; Workman & Hart, 2005.

Rock compositions	Al %	Ti %	Sc ppm	Cr ppm	Zr ppm	Nb ppm	Hf ppm	Ta ppm	Ir ppm
CI chondrite	0.85	0.05	5.90	2646.00	3.86	0.25	0.11	0.01	0.48
CV chondrite	1.68	0.09	10.20	3480.00	8.90	0.50	0.23	0.03	0.73
Primitive Mantle	2.38	0.13	16.50	2520.00	10.81	0.59	0.30	0.04	0.00
DMM	2.30	0.08	16.00	2500.00	8.00	0.20	0.20	0.01	0.00
Ocean Island Basalt	7.67	1.40	30.00	450.00	150.00	14.00	4.10	1.40	0.00
Plateau Basalt	7.32	1.00	54.50	18.00	55.00	2.70	1.70	0.50	0.00
N-MORB	8.63	0.67	44.00	251.00	57.00	1.07	2.90	0.10	0.00
T-MORB	8.51	1.03	43.00	251.00	100.00	5.15	2.12	0.38	0.00
Continental crust	8.42	0.42	21.90	135.00	132.00	8.00	3.70	0.70	0.00
TTG	8.20	0.20	4.70	29.00	152.00	6.40	4.50	0.71	0.00
Apex Basalt - High-Ti	6.55	0.56	38.00	496.00	64.55	3.20	1.96	0.13	0.00
Euro Basalt - High-Ti	6.89	0.61	30.33	475.67	71.72	3.87	2.03	0.12	0.00
Euro Basalt - Low-Ti	7.16	0.36	35.43	474.29	37.36	1.59	1.14	0.03	0.00
Mt Ada Basalt - High-Ti	6.20	0.62	35.82	352.64	78.79	4.02	1.65	0.16	0.00
Mt Ada Basalt - Low-Ti	6.86	0.37	39.00	602.20	48.20	2.12	0.50	0.00	0.00
Cooneeina basalt	7.53	0.30	38.38	247.00	52.03	2.19	1.59	0.14	0.00
Komatiite	1.89	0.18	15.75	1997.81	36.69	1.68	0.72	0.14	0.00
S3 average	7.85	0.58	35.00	966.28	47.46	1.20	1.42	0.07	0.11
Loop S3	6.67	0.50	28.00	1125.00	42.00	1.00	1.26	0.06	0.11

Table DR1: Endmember data for model and averaged data for all three sections of S3 and for the Loop Road section. Data for endmembers from Arevalo and Mcdonough, 2010; Ash et al., 2005; Kemp and Hawkesworth, 2003; Kerr, 2003; Klein, 2003; Lodders and Fegley, 1998; Maier et al., 2005; Palme and Jones, 2003; Palme and O'Neill, 2003; Rudnick and Gao, 2003; Salters and Stracke, 2004; Smithies et al., 2005; Workman and Hart, 2005.