Procedure for outlier detection

Calibration Data Set 1, CDS1. This data set includes all observations but seven outliers (see Fig. 1SC) that appear with either expressions A1 or B1. Outliers were progressively detected by repeating the regression calculations to retrieve the parameters for expression 9. Regression methods based on multiple maximum-likelihood estimators (MM estimators, Yohai, 1987) were used in the calculations.

- **Step 1:**

  Observations used in the calculations: 203, i.e., all compositional pairs.

  Retrieved expression: A1-1 (Table 1SC).

  Excess Gibbs free energy of reaction for plagioclase: $\Delta G_{pl}^{ex}(EG90)$ (Tables 1SC).

  Outlying data: or_00446 and or_00457 (Fig. 1SC; see Appendix A for details).

- **Step 2:**

  Observations used in the calculations: 201; two outlying data excluded.

  Retrieved expression: A1-2.

  Excess Gibbs free energy of reaction for plagioclase: $\Delta G_{pl}^{ex}(EG90)$.

  Outlying data: or_00385.

- **Step 3:**

  Observations used in the calculations: 200; three outlying data excluded.

  Retrieved expression: A1-3.

  Excess Gibbs free energy of reaction for plagioclase: $\Delta G_{pl}^{ex}(EG90)$.

  Outlying data: or_00477 and or_00510.

- **Step 4:**

  Observations used in the calculations: 198; five outlying data excluded.

  Retrieved expression: A1-4.

  Excess Gibbs free energy of reaction for plagioclase: $\Delta G_{pl}^{ex}(EG90)$.

  Outlying data: No outlying data detected.

- **Step 5:**

  Observations used in the calculations: 198; five outlying data excluded.

  Retrieved expression: B1-1.

  Excess Gibbs free energy of reaction for plagioclase: $\Delta G_{pl}^{ex}(HP92)$ (Table 1SC).

  Outlying data: or_00445.

- **Step 6:**

  Observations used in the calculations: 197; six outlying data excluded.

  Retrieved expression: B1-2.

Excess Gibbs free energy of reaction for plagioclase: $\Delta G_{pl}^{ex}(HP92)$.

Outlying data: or_00511.

Definitive expressions A1 and B1 were obtained after excluding the seven outlying data (see values of the fitted parameters for these expressions in Table 5).

Calibration Data Set 2, CDS2. This includes approximately one half of the total data set. Complete data subsets from papers that are consistent with the models found with CDS1 (see Appendixes A) were selected for the calculations. In the first step, the seven outlying data detected in the previous calculations were also included to evaluate their behavior when using a different data set.

- **Step 1:**

  Observations used in the calculations: 104; 12 outlying data included.

  Retrieved expression: A2-1 (Table 2SC).

  Excess Gibbs free energy of reaction for plagioclase: $\Delta G_{pl}^{ex}(EG90)$ (Table 2SC).

  Outlying data: or_00385, or_00445, or_00446, or_00456, or_00457, or_00460, or_00477, or_00508, or_00510 and or_00511 (Fig. 2SC; see Appendix A for details).

- **Step 2:**

  Observations used in the calculations: 94; 10 outlying data excluded.

  Retrieved expression: A2-2.

  Excess Gibbs free energy of reaction for plagioclase: $\Delta G_{pl}^{ex}(EG90)$.

  Outlying data: or_00092 and or_00095.

Definitive expressions A2 and B2 were obtained after excluding the 12 outlying data (see values of the fitted parameters for these expressions in Table 5).

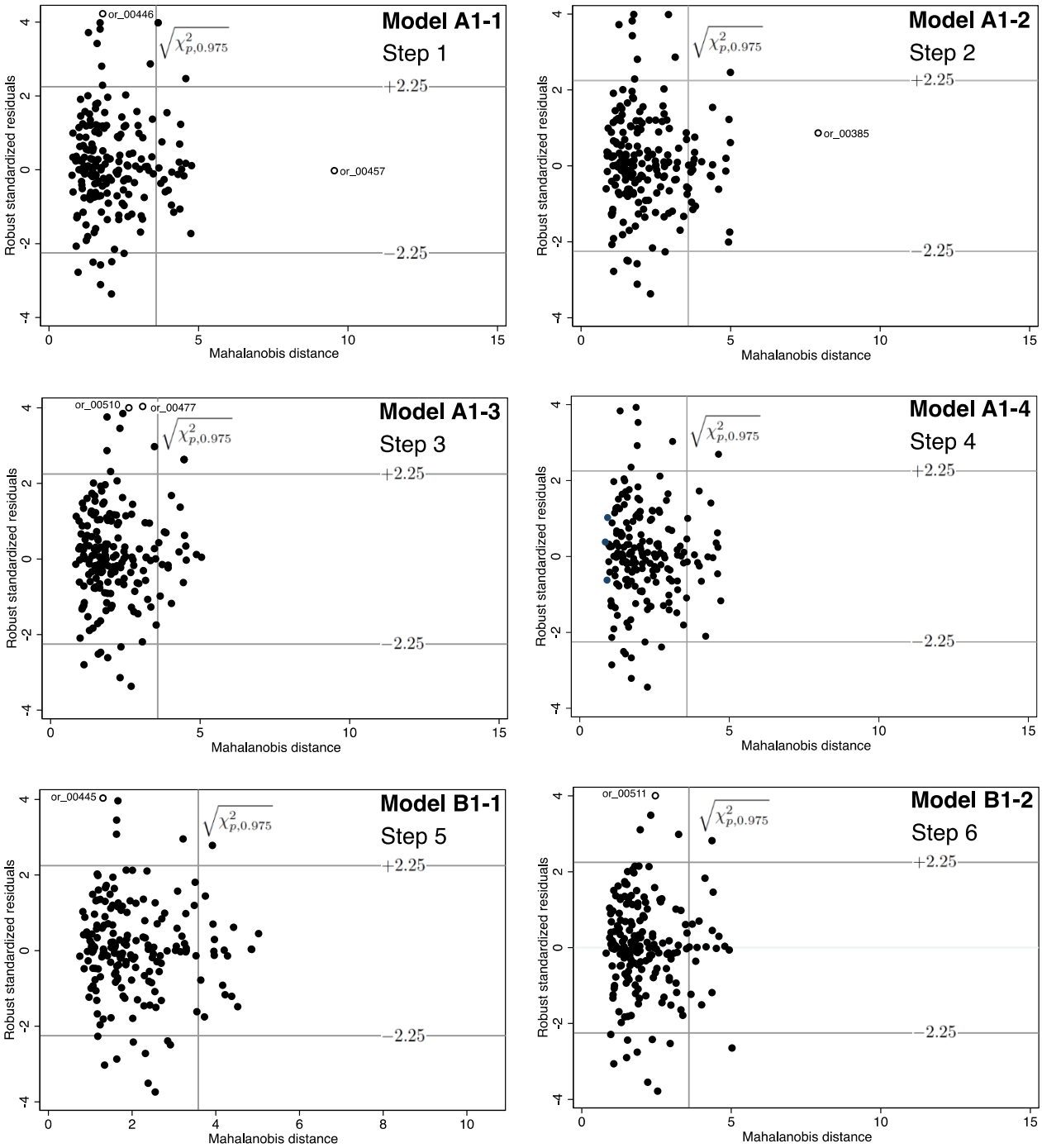

Fig. 1SC. Diagnostic plots of robust standardized residuals versus Mahalanobis distance for the expressions derived in this work using MM-estimators reported in Table 1SC. Thermodynamic models (see text for details): A1-1, calculated with 203 observations and $\Delta G_{pl}^{ex}(EG90)$ (Table 1SC); A1-2, calculated with 201 observations and $\Delta G_{pl}^{ex}(EG90)$; A1-3, calculated with 200 observations and $\Delta G_{pl}^{ex}(EG90)$; A1-4, calculated with 198 observations and $\Delta G_{pl}^{ex}(EG90)$; B1-1, calculated with 198 observations and $\Delta G_{pl}^{ex}(HP92)$ (Table 1SC); and B1-2, calculated with 197 observations and $\Delta G_{pl}^{ex}(HP92)$.

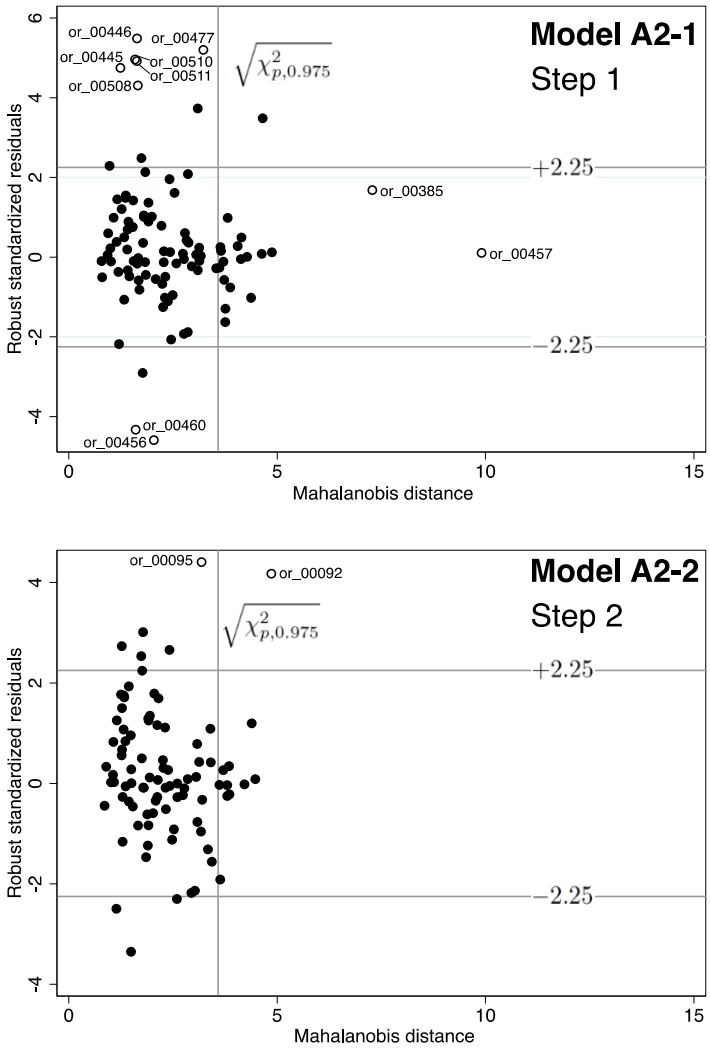

Fig. 2SC. Diagnostic plots of robust standardized residuals versus Mahalanobis distance for the expressions derived in this work using MM-estimators reported in Table 2SC. Thermodynamic models (see text for details): A2-1, calculated with 104 observations and $\Delta G_{pl}^{ex}(EG90)$ (Table 2SC); and A2-2, calculated with 94 observations and $\Delta G_{pl}^{ex}(EG90)$.

Table 1SC. Fitted thermodynamic parameters for reaction *R1* and for $\Delta G^{ex}_{amp} = RTln\gamma_{gln} - RTln\gamma_{ts}$. Modeled expression: $-\left(RTlnK^{id} + \Delta G^{ex}_{pl}\right) = A - TB + PC + \sum_{i=1}^{8}\{p_i W_i^H - p_i TW_i^S + p_i PW_i^V\}$. Parameters retrieved using MM-estimators. Units: J and J/K

Expression	*A1-1*	*A1-2*	*A1-3*	*A1-4*	*B1-1*	*B1-2*
Plagioclase mixing model[1]	$\Delta G^{ex}_{pl}(EG90)$	$\Delta G^{ex}_{pl}(EG90)$	$\Delta G^{ex}_{pl}(EG90)$	$\Delta G^{ex}_{pl}(EG90)$	$\Delta G^{ex}_{pl}(HP92)$	$\Delta G^{ex}_{pl}(HP92)$
Calibration data set[2]	*CDS1+7OUT*	*CDS1+5OUT*	*CDS1+4OUT*	*CDS1+2OUT*	*CDS1+2OUT*	*CDS1+1OUT*
Step	*1º*	*2º*	*3º*	*4º*	*5º*	*6º*
Observations	203	201	200	198	198	197
Scale parameter	6611	6611	6579	6470	6463	6406
Thermodynamic parameters						
A	(1.32±0.23) 10^5 t = 5.7 *P(>|t|)* = 0	(1.32±0.23) 10^5 t = 5.7 *P(>|t|)* = 0	(1.34±0.21) 10^5 t = 6.4 *P(>|t|)* = 0	(1.35±0.21) 10^5 t = 6.4 *P(>|t|)* = 0	(1.39±0.22) 10^5 t = 6.4 *P(>|t|)* = 0	(1.39±0.21) 10^5 t = 6.6 *P(>|t|)* = 0
B	86±20 t = 4.3 *P(>|t|)* = 0	87±21 t = 4.1 *P(>|t|)* = 0	87±20 t = 4.4 *P(>|t|)* = 0	88±20 t = 4.4 *P(>|t|)* = 0	90±22 t = 4.1 *P(>|t|)* = 0	91±21 t = 4.3 *P(>|t|)* = 0
C	0	0	0	0	0	0
W_2^H	(-2.55±0.85) 10^4 t = -3 *P(>|t|)* = 0.003	(-2.54±0.88) 10^4 t = -2.9 *P(>|t|)* = 0.004	(-2.75±0.63) 10^4 t = -4.4 *P(>|t|)* = 0	(-2.78±0.58) 10^4 t = -4.8 *P(>|t|)* = 0	(-3.00±0.55) 10^4 t = -5.5 *P(>|t|)* = 0	(-3.01±0.52) 10^4 t = -5.8 *P(>|t|)* = 0
W_5^H	(7.26±0.75) 10^4 t = 9.7 *P(>|t|)* = 0	(7.27±0.79) 10^4 t = 9.2 *P(>|t|)* = 0	(7.27±0.79) 10^4 t = 9.2 *P(>|t|)* = 0	(7.29±0.79) 10^4 t = 9.2 *P(>|t|)* = 0	(7.04±0.79) 10^4 t = 8.9 *P(>|t|)* = 0	(7.06±0.78) 10^4 t = 9.1 *P(>|t|)* = 0
W_7^S	88±28 t = 3.1 *P(>|t|)* = 0.002	86±45 t = 1.9 *P(>|t|)* = 0.058	100±29 t = 3.4 *P(>|t|)* = 0.001	101±28 t = 3.6 *P(>|t|)* = 0	109±27 t = 4.0 *P(>|t|)* = 0	109±26 t = 4.2 *P(>|t|)* = 0
W_8^S	66±24 t = 2.8 *P(>|t|)* = 0.006	66±24 t =2.8 *P(>|t|)* = 0.007	70±19 t = 3.7 *P(>|t|)* = 0	71±18 t = 3.9 *P(>|t|)* = 0	69±16 t = 4.3 *P(>|t|)* = 0	70±16 t = 4.4 *P(>|t|)* = 0

Notes: 1: $\Delta G^{ex}_{pl}(EG90)$, ternary feldspar solution model of Elkins and Grove (1990); $\Delta G^{ex}_{pl}(HP92)$, DQF approach for plagioclase activity-composition relations of Holland and Powell (1992). 2: CDS1, Calibration Data Set 1; OUT, set of outlying observations excluded for retrieving expressions A1 and B1 (total number of outlying observations: 7); n OUT, number n of observations included in the calculations.

Table 2SC. Fitted thermodynamic parameters for reaction *R1* and for $\Delta G^{ex}_{amp} = RTln\gamma_{gln} - RTln\gamma_{ts}$. Modeled expression: $-\left(RTlnK^{id} + \Delta G^{ex}_{pl}(EG90)\right) = A - TB + PC + \sum_{i=1}^{8}\{p_i W_i^H - p_i T W_i^S + p_i P W_i^V\}$. Parameters retrieved using MM-estimators. Units: J and J/K

Expression	*A2-1*	*A2-2*
Plagioclase mixing model[1]	$\Delta G^{ex}_{pl}(EG90)$	$\Delta G^{ex}_{pl}(EG90)$
Calibration data set[2]	*CDS2+12OUT*	*CDS2+2OUT*
Step	*1º*	*2º*
Observations	104	94
Scale parameter	5407	4659
Thermodynamic parameters		
A	(1.469±0.077) 10_5 t = 19 *P(>|t|)* = 0	(1.474±0.077) 10_5 t = 19 *P(>|t|)* = 0
B	91.3±9.8 t = 9.3 *P(>|t|)* = 0	93±12 t = 7.8 *P(>|t|)* = 0
C	0	0
W_2^H	(-4.00±0.73) 10_4 t = -5.5 *P(>|t|)* = 0	(-3.83±0.72) 10_4 t = -5.3 *P(>|t|)* = 0
W_5^H	(6.59±0.76) 10_4 t = 8.7 *P(>|t|)* = 0	(6.88±1.11) 10_4 t = 6.2 *P(>|t|)* = 0
W_7^S	133±20 t = 6.7 *P(>|t|)* = 0	134±28 t = 4.8 *P(>|t|)* = 0
W_8^S	102±16 t = 6.4 *P(>|t|)* = 0	97±18 t =5.4 *P(>|t|)* = 0

Notes: 1: $\Delta G^{ex}_{pl}(EG90)$, ternary feldspar solution model of Elkins and Grove (1990). 2: CDS2, Calibration Data Set 2; OUT, set of outlying observations excluded for retrieving expressions A2 and B2 (total number of outlying observations: 12); n OUT, number n of observations included in the calculations.

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Holland T.J.B., and Powell R. (1992) Plagioclase feldspars: activity-composition relations based upon Darken's Quadratic Formalism and Landau theory. American Mineralogist, 77, 53–61

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