APPENDIX 2: MIMiC – MELT INCLUSION MODIFICATION CORRECTION

A2.1. Overview

The Melt Inclusion Modification Correction (MIMiC) program corrects melt inclusions for post-entrapment crystallization/melting (PEC/PEM), and it includes optional corrections for Fe-Mg exchange with the host and vapor bubble growth. Here we describe these functions and discuss how to use the program. Two versions of the program are included, one for Python 2 and another for Python 3.

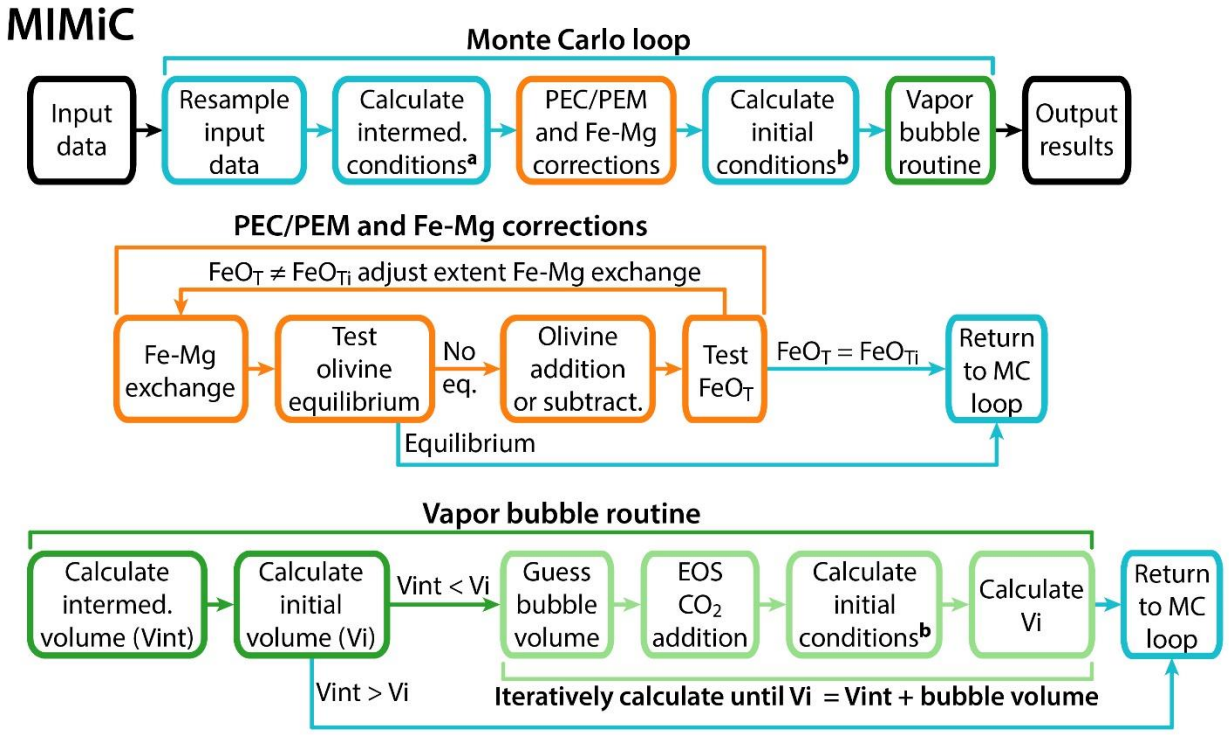

Figure A2.1. Overview of MIMiC. *Intermediate* conditions[a] are the closure temperature for CO_2, calculated based on the cooling rate and water content of the melt, and the intermediate pressure, determined using the volatile contents of the glass. *Initial* conditions[b] are the initial temperature, calculated using the PEC/PEM-corrected melt inclusion composition and the initial pressure, and the initial pressure, determined using the initial volatile contents of the melt and initial temperature. See Appendix 1 for a detailed description of all reconstruction methods.

A2.1.1. Olivine crystallization/melting correction

To address PEC and PEM, MIMiC incrementally adds or subtracts (respectively) equilibrium olivine from the composition of the melt inclusion until equilibrium with the host is reached (e.g., Trial 1 in Figs. A2.2a, A2.2b). The olivine-melt equilibrium expression ($Kd_{Fe\text{-}Mg}^{olv\text{-}melt}$) is calculated using the Toplis (2005) or Ford et al. (1983) model. Both models incorporate the effects of pressure and temperature. Pressure is calculated using VolatileCalc (Newman and Lowenstern, 2002). Temperature is calculated using the thermometer (eq. 4) of Putirka et al. (2007). Upon initiating a correction, host-melt equilibrium is tested using a calculated Kd. If the melt inclusion is found to be in equilibrium with an olivine of higher Fo than that of the host,

equilibrium olivine is removed stepwise (calculating new values of Kd at each step) until equilibrium is reached. Conversely, if the melt inclusion is in equilibrium with a lower Fo olivine than that of the host, equilibrium olivine is added in steps. The incorporation of pressure, stepwise calculation of Kd, and Monte Carlo approach mark significant advantages of our approach relative to others (e.g., Danyushevsky and Plechov, 2011). FeO and Fe_2O_3 are calculated from the user input $Fe^{3+}/\sum Fe$ at the start of the correction. By default, Fe_2O_3 is treated as incompatible during PEC/PEM and Fe-Mg corrections, which is not necessarily the case. We suggest this simplification is of minor significance because incompatible element shifts due to PEC/PEM are typically small (≤5%). Additionally, fO_2 is thought to re-equilibrate at the relatively slow metal vacancy rate (Gaetani et al., 2012; Faul et al., 2018), meaning that over short timescales Fe_2O_3 may behave incompatibly. MIMiC also enables users to hold $Fe^{3+}/\sum Fe$ constant during corrections, which is appropriate to do if Fe speciation equilibrates with the external melt over the timescale of post-entrapment crystallization.

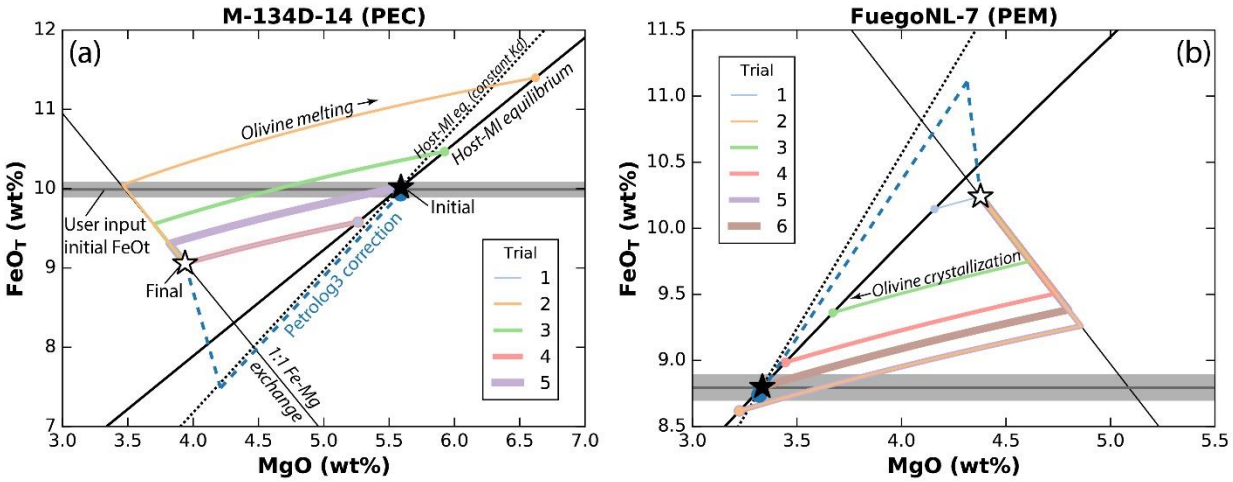

Fig. A2.2. Iterative method used in MIMiC to correct for post-entrapment crystallization (PEC) or melting (PEM) and Fe-Mg exchange (from main text). Examples are shown for two melt inclusions from Fuego. The open star (*Final*) shows the measured melt inclusion composition, and the filled star (*Initial*) is the corrected composition. The gray horizontal line shows the user-defined value of initial FeO_{Ti}, with the range of values that MIMiC accepts (shaded region). Each trial consists of an Fe-Mg exchange correction, which is omitted in the first trial, followed by a PEC or PEM correction. The amount of Fe-Mg exchange is adjusted in each trial until the PEC/PEM correction yields the input value of FeO_{Ti}. The host-inclusion equilibrium (solid black line) is different than the host-inclusion equilibrium at constant Kd (dashed black line) because temperature and melt composition of host-melt equilibrium vary, which affect Kd. Corrections performed using *Petrolog3* (Danyushevsky and Plechov, 2011) are shown with blue, dashed lines.

A2.1.2. Fe-Mg Exchange

Fe-Mg exchange describes the process of Fe-loss from a melt inclusion following PEC by Fe-Mg exchange with the host. The opposite occurs for PEM ("Fe-gain"). See Danyushevsky et al. (2000) for an in-depth examination. Fe-Mg exchange is often identified by finding FeO_T contents of PEC/PEM-corrected melt inclusions higher ("Fe-gain") or lower ("Fe-loss") than those along a representative liquid line of descent or by observing Fo gradients near melt inclusions (Danyushevsky et al., 2000; Gaetani and Watson, 2000). If such evidence exists, MIMiC users can

enable an Fe-Mg exchange correction (Fig. A2.2). FeO_{Ti} contents of the melt inclusions must be input, which can be determined by evaluating Fo-FeO_T trends (Fig. A2.3). Initially, each melt inclusion is corrected for PEC/PEM. The corrected FeO_T contents are checked against the input FeO_{Ti}. If the two values are not within 0.1 wt%, an iterative Fe-Mg correction is initiated. Each iteration involves two steps. First, MIMiC simulates 1:1 molar exchange of Fe-Mg with the host. FeO is increased or decreased (as necessary), and MgO vice versa, by an increment. Second, the PEC/PEM correction is performed. The resultant FeO_T is compared with FeO_{Ti}. If the two are not within 0.1 wt%, another calculation is initiated adjusting the Fe-Mg increment as necessary. This approach is different than that of Petrolog3 (Danyushevsky and Plechov, 2011). Their correction also occurs in two steps. First, concurrent Fe-Mg exchange and PEC/PEM corrections occur at approximately fixed MgO content until equilibrium with the host is reached. Second, FeO_T is set to the user input value, and PEC/PEM and Fe-Mg exchange are simulated in order to maintain olivine-melt equilibrium. MIMiC and Petrolog3 yield different results because Petrolog3 calculates a single value of Kd at the start of each step and does not include the effect of pressure. Alternatively, MIMiC recalculates Kd after each increment of olivine addition, or Fe-Mg exchange, and incorporates the effect of pressure. If we pick a Kd model in Petrolog3 that has the same final Kd as MIMiC calculates, the results agree (Fig. A2.2).

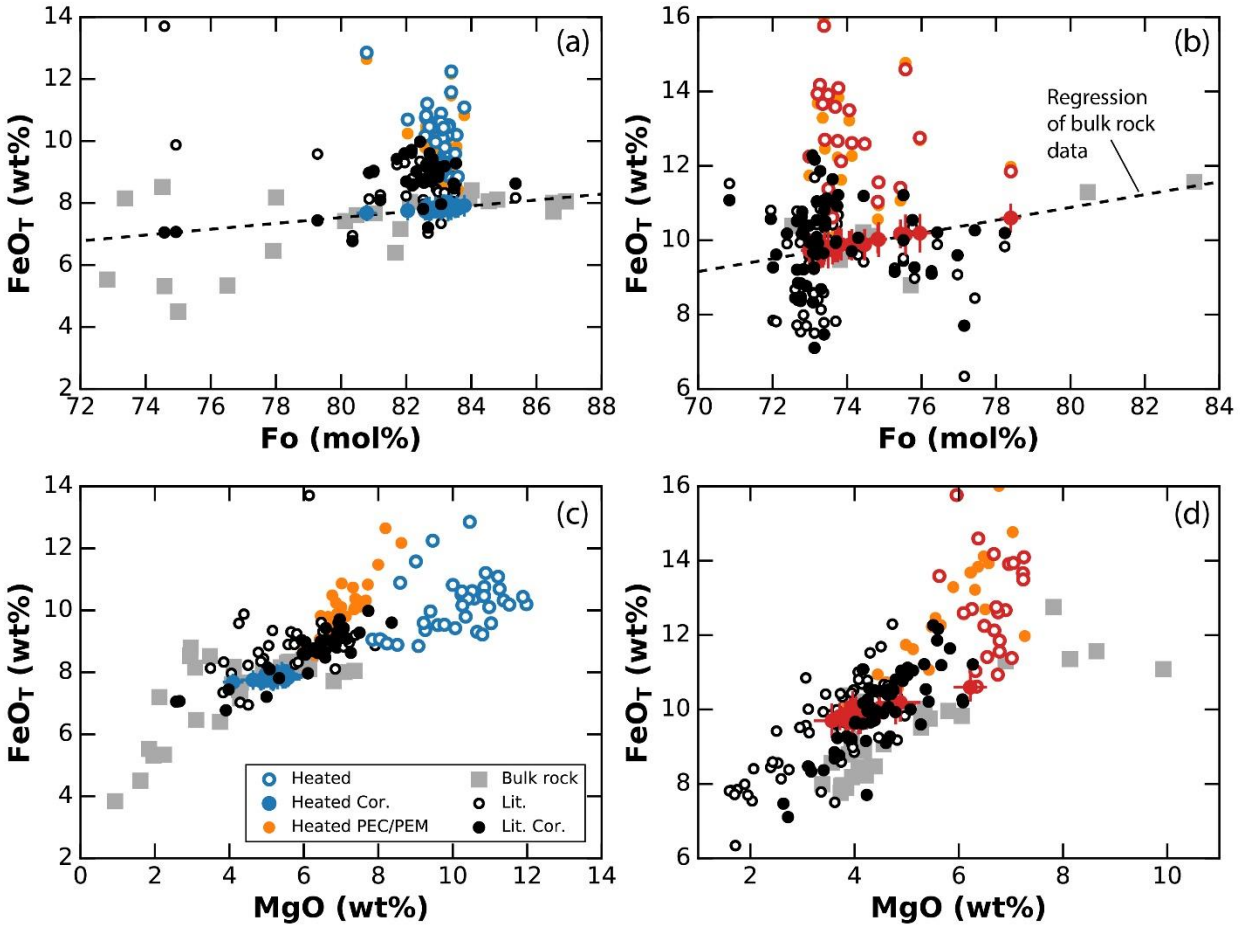

Fig. A2.3. Melt inclusion and bulk rock Fe and Mg contents. In **(a)** and **(b)**, Fo is olivine forsterite content of the host (for melt inclusions) or the equilibrium olivine (bulk rocks). The dashed black lines in **(a)** and **(b)** are a regression of the bulk rock data that is used to determine the initial FeO_T

contents of melt inclusions for Fe-Mg exchange corrections. Open symbols are the measured values. "PEC/PEM" indicates values that have been corrected for PEC/PEM only. "Cor." indicates melt inclusions have been corrected for PEC/PEM and Fe-Mg exchange, when necessary. Note "Heated PEC/PEM" values show very high Fe contents, indicating Fe-gain occurred.

A2.1.3. Vapor bubble correction

The vapor bubble corrections occur after the PEC/PEM corrections are complete. The observed volume and two calculated volume approaches (Riker, 2005; this study) are employed. Details of how these corrections are performed are found in Appendix 1.

A2.1.4. Quantifying uncertainties

A Monte Carlo approach is taken for the corrections. For each MI, *n* corrections are conducted. The first correction is performed on the input variables. In each subsequent correction, the input variables are resampled according to their uncertainties assuming a normal distribution) and the corrections are performed using the resampled data. After *n* calculations are complete, the results of the initial correction are output, and uncertainty is calculated as the standard deviation of the results. Prior to calculating the standard deviation, the modified-Z-score method (see https://www.itl.nist.gov/div898/handbook/eda/section3/eda35h.htm) is used to remove outliers. If *n* is set to 1, the reported uncertainties are those input by the user.

The default value for *n* is 50 (Fig. A2.4).

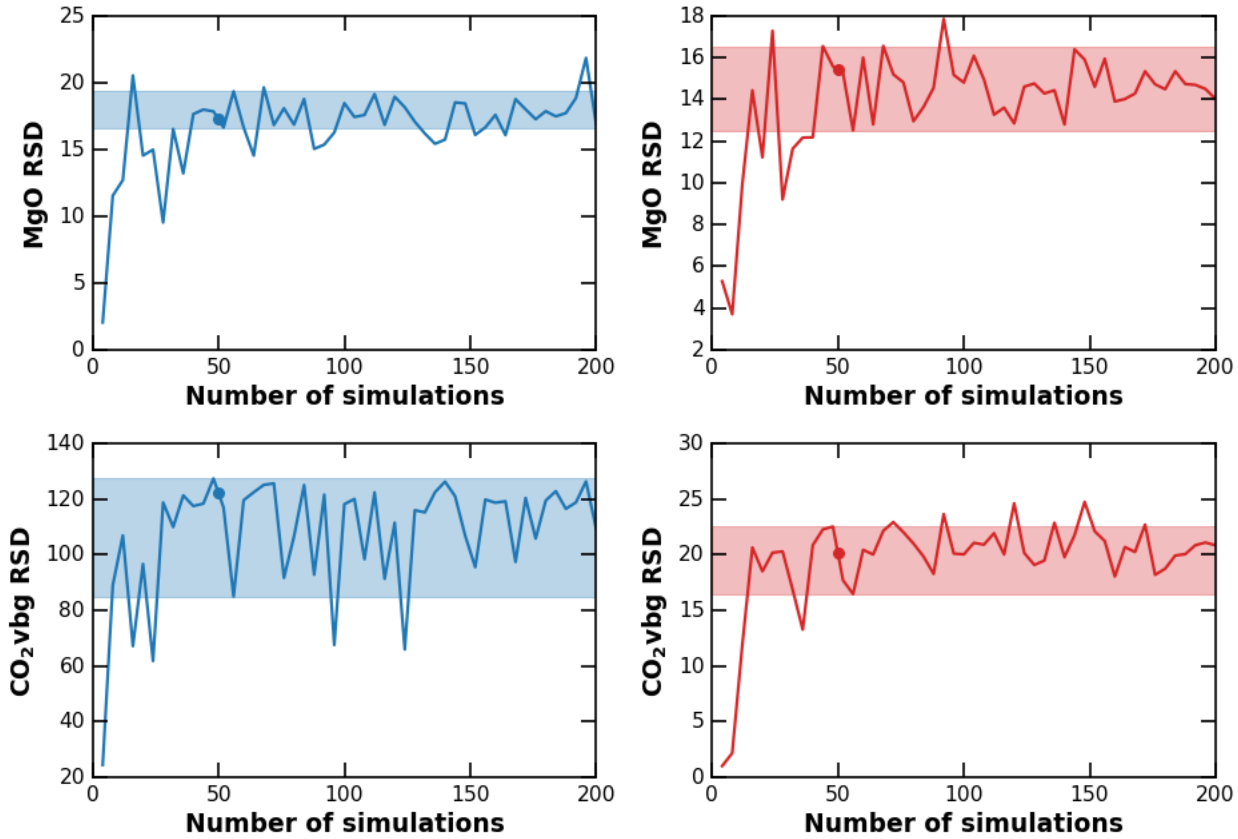

Figure A2.4. Uncertainties resulting from different numbers of Monte Carlo simulations (*n*), which are multiples of 4 up to 200. The left panels are corrections for Seguam melt inclusion *seg 8.3* and the right panels are for Fuego melt inclusion *137B-R-1*. As *n* increases, the resultant relative standard deviation (RSD) values stabilize. Ideally, a large number of corrections ($n > 100$) would produce the most stable results. However, that is computationally expensive, and we recommend a value of *n* of 50, which is indicated which a circle. The shaded region shows the maximum and minimum RSD values from trials with *n* equal to 44, 48, 52, and 56.

A2.1.5. Models employed

MIMiC makes use of several previously published models. Cite each appropriately.

Fe-Mg olivine-melt partitioning ($K_D^{Fe\text{-}Mg}$): The user can choose between two K_D models that are provided: Ford et al. (1983) and Toplis (2005).

Ford, C.E., Russell, D.G., Craven, J.A., and Fisk, M.R. (1983) Olivine-Liquid Equilibria: Temperature, Pressure and Composition Dependence of the Crystal/Liquid Cation Partition Coefficients for Mg, Fe2+, Ca and Mn. Journal of Petrology, 24(3), 256-266.

Toplis, M.J. (2005) The thermodynamics of iron and magnesium partitioning between olivine and liquid: criteria for assessing and predicting equilibrium in natural and experimental systems. Contributions to Mineralogy and Petrology, 149(1), 22-39.

Thermometry: MIMiC uses the olivine-melt model (eq. 4) of Putirka et al. (2007). In this calculation we use FeO_T for FeO.

Putirka, K.D., Perfit, M., Ryerson, F.J., and Jackson, M.G. (2007) Ambient and excess mantle temperatures, olivine thermometry, and active vs. passive upwelling. Chemical Geology, 241(3–4), 177-206.

Barometry: H_2O-CO_2 solubility is described by VolatileCalc (Newman and Lowenstern, 2002).

Newman, S., and Lowenstern, J.B. (2002) VolatileCalc: a silicate melt–H_2O–CO_2 solution model written in Visual Basic for excel. Computers & Geosciences, 28(5), 597-604.

Volume calculations and density: MIMiC uses the compilation of Lesher and Spera (2015).

Lesher, C.E., and Spera, F.J. (2015) Thermodynamic and transport properties of silicate melts and magma. The Encyclopedia of Volcanoes (Second Edition), p. 113-141. Elsevier.

Olivine density

Hacker, B.R., Peacock, S.M., Abers, G.A., and Holloway, S.D. (2003) Subduction factory 2. Are intermediate-depth earthquakes in subducting slabs linked to metamorphic dehydration reactions? Journal of Geophysical Research: Solid Earth, 108(B1).

Olivine thermal expansion coefficients

Liu, W., and Li, B. (2006) Thermal equation of state of (Mg0.9Fe0.1)2SiO4 olivine. Physics of the Earth and Planetary Interiors, 157(3), 188-195.

Closure temperatures: The formulation of Dodson (1973) is used.

Dodson, M.H. (1973) Closure temperature in cooling geochronological and petrological systems. Contributions to Mineralogy and Petrology, 40(3), 259-274.

Glass transition temperatures: The viscosity threshold is set using the calculation of Zhang et al. (2007) and viscosity is calculated using the model of Giordano et al. (2008).

Zhang, Y., Xu, Z., Zhu, M., and Wang, H. (2007) Silicate melt properties and volcanic eruptions. Reviews of Geophysics, 45(4).
Giordano, D., Russell, J.K., and Dingwell, D.B. (2008) Viscosity of magmatic liquids: a model. Earth and Planetary Science Letters, 271(1-4), 123-134.

Calculated volume approach: Two calculated volume approaches are employed. One is the new vapor bubble growth model presented in this paper. The other is the model of Riker (2005).

Riker, J. (2005) The 1859 Eruption of Mauna Loa Volcano, Hawai'i: Controls on the Development of Long Lava Channels. University of Oregon.

A2.2. Instructions for using MIMiC

A2.2.1. Requirements

The code can be run in Python 2 or 3. The following packages are required:

- *csv*
- *numpy*
- *scipy*

The user must change the name of "Appendix2.2_MIMiC_mi_functions_Py2_V1.py" to "mi_functions_Py2_V1.py" (if using Python 2) or "Appendix2.2_MIMiC_mi_functions_Py3_V1.py" to "mi_functions_Py3_V1.py" (if using Python 3) .

A2.2.2. User input data

Use "Appendix2.4_ExampleInputData.csv" as a template for data input. The file is also an example. The output of this file is shown in "Appendix2.5_ExampleOutput.csv". Notes on the input parameters are given below.

As noted in the main text, if estimates for cooling rate or melt inclusion and host radius are not available, MIMiC estimates one or the other. If cooling rate estimates do not exist, cooling rate is calculated by MIMiC using the Dodson equation (eq. 2, main text) rearranged to solve for the cooling rate (τ). In this case, known values are required for T_c (closure temperature), E_A (activation energy), R (gas constant), A (geometry constant), D_0 (diffusivity at infinite temperature), and a (melt inclusion radius). We assume T_c (MgO) is equivalent to the temperature calculated using the equilibrium olivine and measured melt inclusion composition (T_{eqolv}). We tested this assumption by comparing T_c (MgO), known value of τ, and T_{eqolv} in heated melt inclusions, and the two agree within 5%. The assumption is not valid if MgO diffusion has not reached the center of the melt inclusion. If no morphological data exist, the host radius is assumed to be 0.5 mm. The radius of the melt inclusion (r_{MI}) is calculated by MIMiC using the Dodson equation (eq. 2) in a similar manner to the cooling rate approach. In this case, the equation is rearranged to solve for the diffusion length (a). Known values are required for T_c, E_A, R, A, D_0, and τ. Therefore, MIMiC can be performed without one of cooling rate or morphological (i.e., melt inclusion and host radii) data.

SAMPLE
Sample identifier.

SIO2, TIO2, AL2O3, FEOT, MNO, MGO, CAO, NA2O, K2O, P2O5, S, CL, H2O, CO2

Compositional data. All values are in wt%, except S, CL, and CO2, which are in ppm (by weight). If any of these fields are left blank, a value of zero is assumed. If H_2O is left blank or set to zero, the calculations will be performed assuming a pressure of 1 MPa.

FO
Forsterite content of the host, given as a percent (i.e., a value between 0-100). Only values between 0 and 100 are accepted.

FE3FET
Fe^{3+}/Fe^{T} of the melt, given as a fraction (i.e., a value between 0-1). If no value (or a value of zero) is entered, a value of 0.2 is assumed.

FEOTI
Value of initial FeOT contents of the melt inclusion (in wt%). Used only in Fe-Mg corrections. If no value (or a value of zero) is entered, no Fe-Mg correction will be performed.

VBVOLP
Vapor bubble volume percent (i.e., a value between 0-100).

DMI
Diameter of the melt inclusion (in µm). If vapor bubble corrections are performed and no value (or a value of zero) is entered, *DMI* will be calculated using the method outlined above. The minimum accepted value is 5 µm and the maximum is 300 µm.

DHOST
Diameter of the host olivine (in µm). If vapor bubble corrections are performed and no value (or a value of zero) is entered, a value of 1000 µm will be assumed. DHOST must be equivalent to at least two times DMI and less than 4 mm.

CR
Cooling rate (in °C/s). If no value (or a value of zero) is entered and DMI is provided, cooling rate is calculated by determining the cooling rate at which the calculated MgO closure temperature is equivalent to the temperature calculated based on the measured melt inclusion composition and the equilibrium olivine (Teqolv). If not value (or a value of zero) is entered and DMI is not provided, a value of 10 °C/s is assumed (appropriate for ash). Values of less than 0.001 °C/s are assumed to be equal to 0.001 °C/s.

(Input parameter)err
The 1σ standard deviation of the input parameters described above. If no value (or a value of zero) is entered, a value of zero is assumed.

A2.2.3. Initiating a run

There are several variables the user must define before running the code. They are all in the *User Inputs* section at the top of *MIMiC.py*. This is the only section of that users should modify.

melt_inclusion_file

Enter a string with the path to the input file.

output_file
Enter a string with the path to the output file.

vb_cor
Enter either *0* or *1*.
0 – Do not perform vapor bubble corrections. By default, the correction is skipped if either *H2O* or *CO2* are left blank (or either is set to zero) in the input file.
1 (default) – Perform vapor bubble corrections. *H2O* and *CO2* must be entered in the input file.

fe_cor
Enter either *0* or *1*.
0 (default) – Do not perform Fe-Mg exchange corrections. By default, this correction is skipped if *FEOTI* is left blank in the input file.
1 – Perform Fe-Mg exchange corrections. A value of *FEOTI* must be entered in the input file.

Fe_fixed
Enter either *0* or *1*.
0 (default) – Fe speciation is not fixed during PEC/PEM and Fe-Mg exchange corrections.
1 – Fe speciation is held constant during PEC/PEM and Fe-Mg exchange corrections.

kd_model
Enter either *0* or *1*.
0 (default) – Toplis (2005) is used to calculate Kd.
1 – Ford et al. (1983) is used to calculate Kd.

H2O_diff
Enter either *0* or *1*. For melt inclusions without water data (was left blank, or a value of zero was entered):
0 (default) – Water is assumed to be zero.
1 – Water is calculated by difference.

n
Enter the number of corrections to be performed on each melt inclusion. The input value must be an integer. If *1* is entered, a single correction will be performed. If a number greater than *1* is entered, a Monte Carlo simulation will be performed. The default value of *n* is *50*. If uncertainty does not need to be quantified, the correction can be made much faster by setting *n* equal to *1*.

A2.2.4. Output
The output file contains a number of results.

SAMPLE
Sample identifier.

SIO2, TIO2, AL2O3, FEOT, MNO, MGO, CAO, NA2O, K2O, P2O5, S, CL, H2O, CO2

Results of PEC/PEM, and Fe-Mg if selected, correction(s). The CO_2 reported here is not corrected for vapor bubble growth. The units are the same as in the input file. The data are normalized to their initial sum.

CO2obsvol (if vapor bubble correction is turned on)
Results of observed volume CO_2 reconstruction.

CO2riker (if vapor bubble correction is turned on)
Results of the calculated volume CO_2 reconstruction using the Riker (2005) model.

CO2vbg (if vapor bubble correction is turned on)
Results of the calculated volume CO_2 reconstruction using our new vapor bubble growth model.

FO
Host olivine Fo (mol%).

TcCO2
Closure temperature for CO_2 calculated using the Dodson equation.

TcMGO
Closure temperature for MgO calculated using the Dodson equation.

Teqolv
Olivine-melt equilibrium temperature calculated using eq. 4 of Putirka et al. (2007) with calculated equilibrium olivine and measured melt compositions.

Ti
Initial temperature calculated using eq. 4 of Putirka et al. (2007) with the measured olivine and the corrected (PEC/PEM, Fe-Mg if selected, vapor bubble growth using our reconstruction method if the bubble correction is applied) melt composition.

OLV ADD
The wt% of olivine added (positive value) or subtracted (negative value) to the melt composition during the PEC or PEM, respectively, correction.

KD
Fe-Mg distribution coefficient between olivine and melt calculated using either Toplis (2005) or Ford et al. (1983).

Px, Dx, VBvol x
Pressure (in MPa) calculated using VolatileCalc (Newman and Lowenstern, 2002), depth (in km) calculated using a general density model (Fig. A2.5), and vapor bubble volume (vol%). Pressure and depth results are based on CO_2 contents of the glass (*Pglass*, *Dglass*), reconstructed using the observed volume approach (*Pobsvol*, *Dobsvol*), reconstructed using the model of Riker (2005) (*Priker*, *Driker*), and reconstructed using our new vapor bubble growth model (*Pvbg*, *Dvbg*).

Vapor bubble volumes are reported for the Riker (2005) approach (*VBvol riker*) and the vapor bubble growth model from this paper (*VBvol vbg*).

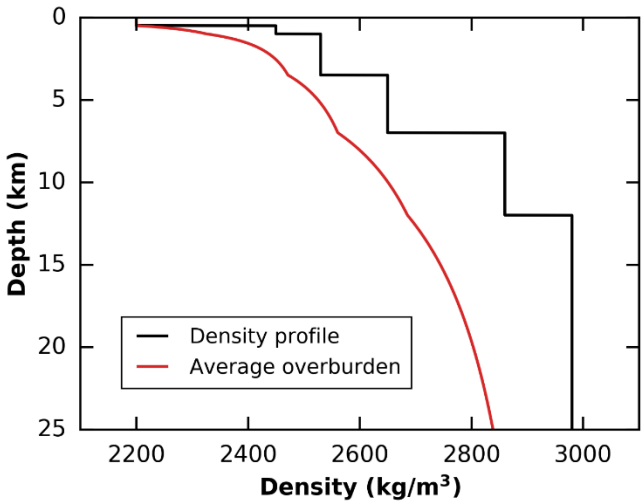

Figure A2.5. Density model derived from a generic seismic velocity profile at Seguam volcano.

Tg (if vapor bubble correction is turned on)
Glass transition temperature (°C) calculated using the method described in Appendix 1.

aMgO (if vapor bubble correction is turned on)
Diffusive length scale for MgO calculated by rearranging the Dodson equation to solve for a. *TcMGO* is assumed to be equivalent to *Teqolv*.

Rmi
Radius of the melt inclusion.

CR
Cooling rate for the melt inclusion.

Initial density (if vapor bubble correction is turned on)
Density (kg/m^3) calculated using the initial melt composition corrected for vapor bubble growth using our new vapor bubble growth model.

Error messages
A record of errors that occurred during corrections. The number of times the error was encountered is indicated by the number in the square brackets following the error message. The messages include the following:

'(Initial or intermediate) SIO2 outside VolatileCalc calibration'
VolatileCalc is calibrated to 40 to 49 wt% SiO_2. If SiO_2 is outside this range, it is assumed to be the nearest value within the range.

'No water information, pressure assumed to be 1 MPa'

If no water data was entered, and water was not calculated by difference, pressure is assumed to be 1 MPa for thermometry and olivine-melt equilibrium calculations.

'(Intermediate, Tg, or Tc CO2) temperature outside VolatileCalc calibration'
VolatileCalc is calibrated to 600 to 1500 ºC. If temperature is outside this range, it is assumed to be the nearest value within the range.

'100% olivine added (too much)'
If 100% of olivine is added during the PEC/PEM correction, the calculation is terminated.

'Could not reach a solution for Fe-Mg correction'
If a solution was not found after 50 iterations, the Fe-Mg correction is terminated.

'Tc (CO2) lower than Tg, Tg used instead'
CO2 diffusion is assumed close at the center of the melt inclusion either at Tc (CO2) or Tg, whichever is higher.

'No solution to bubble growth model found'
If a solution was not found after 50 iterations, the bubble growth correction is terminated.

x1sig
The 1σ standard deviation of Monte Carlo results, if $n > 1$, for all of the outputs. Otherwise, this value is the user input value.

xnorm100
The composition of the corrected melt inclusion normalized to 100%. The CO_2 reported is the PEC/PEM and Fe-Mg, if applicable, corrected glass composition.

xnormANHYD
The composition of the corrected melt inclusion normalized to 100% without volatiles (H_2O, CO_2, S, Cl).

xnormANHYD-FEOT
The composition of the corrected melt inclusion normalized to 100% without volatiles (H_2O, CO_2, S, Cl) and assuming all ferrous Fe.

A2.3. References

Danyushevsky, L.V., Della-Pasqua, F.N., and Sokolov, S. (2000) Re-equilibration of melt inclusions trapped by magnesian olivine phenocrysts from subduction-related magmas: petrological implications. Contributions to Mineralogy and Petrology, 138(1), 68-83.

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Faul, U.H., Cline II, C.J., Berry, A., Jackson, I., and Garapić, G. (2018) Constraints on oxygen fugacity within metal capsules. Physics and Chemistry of Minerals, 45(6), 497-509.
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Gaetani, G.A., O'Leary, J.A., Shimizu, N., Bucholz, C.E., and Newville, M. (2012) Rapid reequilibration of H2O and oxygen fugacity in olivine-hosted melt inclusions. Geology, 40(10), 915-918.
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Zhang, Y., Xu, Z., Zhu, M., and Wang, H. (2007) Silicate melt properties and volcanic eruptions. Reviews of Geophysics, 45(4).