2. METHODS

2.1. Model for calculating volcanic outgassing speciation

Below, we describe our model for predicting the gases produced by an erupting mantle-sourced volcano. We follow the work of Gaillard & Scaillet (2014), and solve for the gas-gas and gas-melt equilibrium in a C-O-H system. Our model differs from Gaillard & Scaillet (2014) because we do not consider nitrogen or sulfur species. Our Python code is published as an open-source software on the Github page https://github.com/Nicholaswogan/Volcano-Speciation.

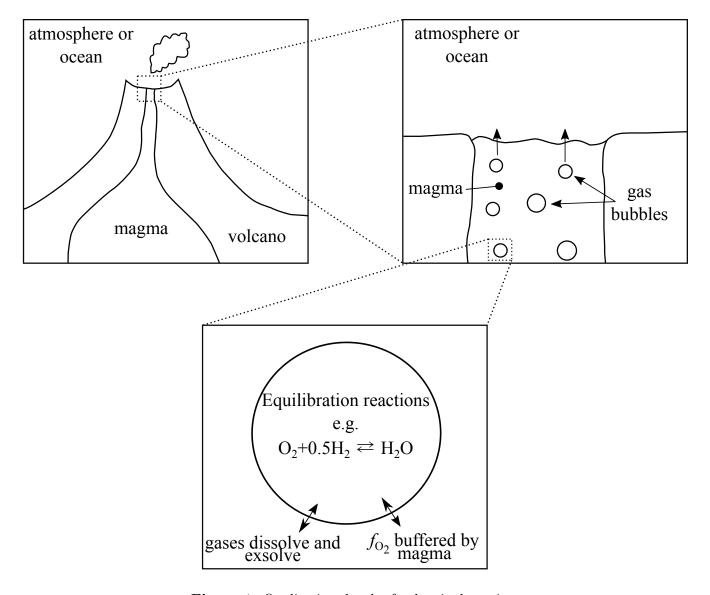


Figure 1. Qualitative sketch of volcanic degassing.

Figure 1 illustrates how we imagine volcanic degassing occurring. Gas bubbles form in the magma when molecules like H_2O and CO_2 are exsolved from the magma. Within the gas bubbles, reactions drive the system to chemical equilibrium. The oxygen fugacity (f_{O_2}) of the gas bubble is enforced by the oxygen fugacity of the magma. Gases bubbles are released from the magma during eruptions and enter the overlying atmosphere or ocean.

Following is our mathematical model describing the volatiles in gas bubbles and in the magma. The amount of carbon and hydrogen that is exsolved by the magma into bubbles is governed by the solubility of CO_2 and H_2O , which we calculate with the solubility relations described in Iacono-Marziano et al. (2012):

$$\ln(x_{\text{CO}_2}) = x_{\text{H}_2\text{O}} d_{\text{H}_2\text{O}} + a_{\text{CO}_2} \ln(P_{\text{CO}_2}) + F_1 \tag{1}$$

$$\ln(x_{\rm H_2O}) = a_{\rm H_2O} \ln(P_{\rm H_2O}) + F_1 \tag{2}$$

Definition Constant Value Units 2.3 Solubility constant $d_{\rm H_2O}$ 1 Solubility constant $a_{\rm CO_2}$ 0.54 Solubility constant $a_{\rm H_2O}$ F_1 -14.23Solubility constant -5.93 F_2 Solubility constant 1.55×10^{-2} $(\text{mol magma})(\text{g magma})^{-1}$ Inverse molar mass of $x_{\rm m}$ magma 18.02 $(g H_2O)(mol H_2O)^{-1}$ Molar mass of H₂O $\mu_{\mathrm{H_2O}}$ $(g CO_2)(mol CO_2)^{-1}$ 44.01 Molar mass of CO₂ $\mu_{\rm CO_2}$ $bar^{0.5}$ Equilibrium constant K_1 $\exp(-29755/T + 6.55)$ $bar^{0.5}$ $\exp(-33979/T + 10.42)$ Equilibrium constant K_2

Table 1: Model constants

 $\frac{K_3}{\text{Note}}$

All constants and variables are defined in Table 1 and Table 2, respectively. The constants F_1 and F_2 depend on the chemical makeup of the magma, which we fix to the concentration of the lava erupting at Mt. Etna, Italy (See Appendix A.1). Using other magma compositions did not significantly change calculated gas speciation. In the equations above, volatile abundance in the magma is expressed as a mol fraction, although this can be converted to a mass fraction with the formula

$$m_i = x_i \mu_i x_{\rm m} \tag{3}$$

Equilibrium constant

Here, m_i is the mass fraction of volatile i (either H_2O or CO_2).

 $\exp(-96444/T + 0.22)$

We assume that after the hot gas exsolves from the magma into bubbles, it achieves thermodynamic equilibrium from the reactions

$$H_2O \leftrightarrow H_2 + \frac{1}{2}O_2$$
 (4)

$$CO_2 \leftrightarrow CO + \frac{1}{2}O_2$$
 (5)

$$CO_2 + 2H_2O \leftrightarrow CH_4 + 2O_2$$
 (6)

At thermodynamic equilibrium, the ratios of the fugacities of volatile species (denoted f_x) are related to the equilibrium constant corresponding to each chemical reaction. We assume that we can replace fugacities with partial pressures (denoted P_x). This approximation is reasonable for the temperatures and pressures involved in volcanic outgassing (Holland 1984).

$$K_1 = \frac{f_{\rm H_2} f_{\rm O_2}^{0.5}}{f_{\rm H_2O}} \approx \frac{P_{\rm H_2} f_{\rm O_2}^{0.5}}{P_{\rm H_2O}} \tag{7}$$

$$K_2 = \frac{f_{\rm CO}f_{\rm O_2}^{0.5}}{f_{\rm CO_2}} \approx \frac{P_{\rm CO}f_{\rm O_2}^{0.5}}{P_{\rm CO_2}} \tag{8}$$

Table 2: Model variables

	Table 2: Model variables		
	Variable	Units	Definition
	P	bar	Total pressure of degassing
Input	T	K	Temperature of magma and gas
	f_{\odot}	bar	of degassing Oxygen fugacity of the magma
	f_{O_2}		
	$m_{ m CO_2}^{ m tot}$	$(g CO_2)(g gas and magma)^{-1}$	mass fraction CO_2 in magma
			before degassing
	$m_{ m H_2O}^{ m tot}$	$(g CO_2)(g gas and magma)^{-1}$	mass fraction H_2O in magma
	-		before degassing
	$x_{ m H_2O}$	$(\text{mol H}_2\text{O}) (\text{mol magma})^{-1}$	mol fraction of H ₂ O in the
			magma after degassing
	$x_{\rm CO_2}$	$(\text{mol CO}_2) \ (\text{mol magma})^{-1}$	mol fraction of CO_2 in the
	_	, , , , , , , , , , , , , , , , , , , ,	magma after degassing
Output	$P_{ m H_2O}$	bar	Partial pressure of H_2O
	P_{CO_2}	bar	Partial pressure of CO ₂
	$P_{ m H_2}$	bar	Partial pressure of H ₂
	P_{CO}	bar	Partial pressure of CO
	$P_{\mathrm{CH_4}}$	bar	Partial pressure of CH_4
	$lpha_{ m gas}$	$(\text{mol gas})(\text{mol gas and magma})^{-1}$	mol fraction in gas phase

$$K_3 = \frac{f_{\text{CH}_4} f_{\text{O}_2}^2}{f_{\text{CO}_2} f_{\text{H}_2\text{O}}^2} \approx \frac{P_{\text{CH}_4} f_{\text{O}_2}^2}{P_{\text{CO}_2} P_{\text{H}_2\text{O}}^2} \tag{9}$$

We calculate equilibrium constants (e.g. K_1) using the NASA thermodynamic database (Burcat & Ruscic 2005). We assume that the gas is thermally and chemically coupled to the magma so that the oxygen fugacity (f_{O_2}) of the gas is set by the oxygen fugacity of magma. Notice that we have 7 unknowns (x_{CO_2} , x_{H_2O} , P_{CO_2} , P_{H_2O} , P_{CO} , P_{H_2} , P_{CH_4}) and only 5 equations. To close the system, we add three more equations and one more unknown. The first equation requires that the partial pressures sum to the total pressure:

$$P_{\rm H_2} + P_{\rm H_2O} + P_{\rm CO} + P_{\rm CO_2} + P_{\rm CH_4} = P \tag{10}$$

The final two equations are atom conservation equations for carbon and hydrogen:

$$\frac{m_{\text{CO}_2}^{\text{tot}}}{\mu_{\text{CO}_2} x_m} = \frac{P_{\text{CO}_2} + P_{\text{CO}} + P_{\text{CH}_4}}{P} \alpha_{\text{gas}} + (1 - \alpha_{\text{gas}}) x_{\text{CO}_2}$$
(11)

$$\frac{m_{\rm H_2O}^{\rm tot}}{\mu_{\rm H_2O}x_m} = \frac{P_{\rm H_2O} + P_{\rm H_2} + 2P_{\rm CH_4}}{P}\alpha_{\rm gas} + (1 - \alpha_{\rm gas})x_{\rm H_2O}$$
(12)

Equations (11) and (12) state that the total moles of either carbon or hydrogen should be equal to the moles of either element in the gas phase plus the moles in the magma. Here, α_{gas} is the final unknown. It is the total moles in the gas phase divided by the total moles in the gas and magma combined. See Appendix A.2 for a full derivation of Equations (11) and (12).

Given a gas/magma temperature (T), pressure (P), oxygen fugacity $(f_{\rm O_2})$, and the total mass fraction (or mol fraction) of ${\rm CO_2}$ and ${\rm H_2O}$ in the magma $(m_{\rm CO_2}^{\rm tot}, {\rm and} \ m_{\rm H_2O}^{\rm tot})$, Equations (1), (2), (7) - (12) are a system of 8 equations and 8 unknowns $(x_{\rm CO_2}, x_{\rm H_2O}, P_{\rm CO_2}, P_{\rm H_2O}, P_{\rm CO}, P_{\rm H_2}, P_{\rm CH_4}, \alpha_{\rm gas})$. We solve this system of equations numerically (see lead author's Github page)

Once the unknowns are solved for, they can be used to calculate the gas production, i.e., the moles of gas produced per kilogram of magma erupted:

$$q_i = \left(\frac{\alpha_{\rm gas} x_{\rm m}}{1 - \alpha_{\rm gas}}\right) \frac{P_i}{P} \tag{13}$$

Here, q_i is the gas production of species i in mol gas/kg magma. Simple unit analysis can be used to confirm Equation (13). Calculating q_i is useful because it is related to the flux of gas i to the atmosphere by the magma production rate:

$$F_i = q_i Q_m \tag{14}$$

Here, Q_m is the magma production rate in kg magma/yr, and F_i is the flux of gas i in mol/yr.

Several authors have shown that degassing can be effected by graphite saturation of magma (Hirschmann & Withers 2008) or by the solubility of CO, CH₄, and H₂ in magma (Wetzel et al. 2013; Ardia et al. 2013; Hirschmann et al. 2012). The gas speciation model described above does not account for these processes although Appendix A.3 shows that our simplified model produces similar results to a model which accounts for graphite saturation and CO, CH₄, and H₂ solubility.

APPENDIX

A. DETAILS OF MAGMA SPECIATION MODEL

A.1. Solubility constants for of H_2O and CO_2

Our outgassing model uses solubility equations for H₂O and CO₂ from Iacono-Marziano et al. (2012) (Equations (1) and (2)). Their solubility equations depend on the chemical make-up of the magma. We found that different magma compositions did not significantly affect magma solubility or the outputs of our outgassing speciation model (Section 2.1), therefore, we fixed the chemical make-up of the magma to the magma erupting at Mt. Etna, Italy. This reduced the complexity of the model without sacrificing any significant amount of accuracy.

Table 4 shows the chemical make-up of the magma at Mt. Etna, and Table 5 shows several solubility constants from Iacono-Marziano et al. (2012). All these values collapse to F_1 and F_2 :

$$F_{1} = \ln(10^{-6}) + \frac{C_{\text{CO}_{2}}P'}{T'} + B_{\text{CO}_{2}} + b_{\text{CO}_{2}} \left[\frac{\text{NBO}}{\text{O}}\right] + \left(\frac{x_{\text{Al}_{2}\text{O}_{3}}}{x_{\text{CaO}} + x_{\text{K}_{2}\text{O}} + x_{\text{Na}_{2}\text{O}}}\right) d_{\text{Al}_{2}\text{O}_{3}/(\text{CaO} + \text{K}_{2}\text{O} + \text{Na}_{2}\text{O})} + (x_{\text{FeO}} + x_{\text{MgO}}) d_{\text{FeO}} + y_{\text{MgO}} + (x_{\text{Na}_{2}\text{O}} + x_{\text{K}_{2}\text{O}}) d_{\text{Na}_{2}\text{O}} + y_{\text{K}_{2}\text{O}} + y_{\text{Na}_{2}\text{O}} + y_{\text{Na}_{2}$$

$$F_2 = \ln(1/(100\mu_{\rm H_2O}x)) + \frac{C_{\rm H_2O}P'}{T'} + B_{\rm H_2O} + b_{\rm H_2O} \left[\frac{\rm NBO}{\rm O}\right]$$
 (A2)

$$\left[\frac{\text{NBO}}{\text{O}}\right] = \frac{2(x_{\text{K}_2\text{O}} + x_{\text{Na}_2\text{O}} + x_{\text{CaO}} + x_{\text{MgO}} + x_{\text{FeO}} - x_{\text{Al}_2\text{O}_3})}{2x_{\text{SiO}_2} + 2x_{\text{TiO}_2} + 3x_{\text{Al}_2\text{O}_3} + x_{\text{MgO}} + x_{\text{FeO}} + x_{\text{CaO}} + x_{\text{Na}_2\text{O}} + x_{\text{K}_2\text{O}}}$$
(A3)

Here, we treat the pressure and Temperature P' and T' as constants with values of 10 bar and 1200 K. This is justified because the terms which depend on pressure and temperature have insignificant contributions to F_1 and F_2 . The result are solubility constants $F_1 = -14.24$ and $F_2 = -5.93$ for CO_2 and H_2O respectfully.

Following is the derivation for the atom conservation equation for carbon used in our outgassing model (Equation (11)). The derivation for the atom conservation equation for hydrogen follows the exact same procedure, so we do not include it.

Table 4: Mt. Etna magma composition

Magma component	Mole fraction	
x_{SiO_2}	0.516	
x_{TiO_2}	0.014	
$x_{\mathrm{Al_2O_3}}$	0.110	
$x_{ m FeO}$	0.091	
$x_{ m MgO}$	0.092	
x_{CaO}	0.126	
$x_{ m Na2O}$	0.035	
$x_{ m K_2O}$	0.002	
$x_{P_2O_5}$	0.016	

Note. Taken from Iacono-Marziano et al. (2012).

Table 5: Solubility constants

Constant	value
$C_{ m CO_2}$	0.14
$B_{ m CO_2}$	-5.3
b_{CO_2}	15.8
$B_{ m H_2O}$	-2.95
$b_{ m H_2O}$	1.24
$d_{\mathrm{Al_2O_3/(CaO+K_2O+Na_2O)}}$	3.8
$d_{ m FeO+MgO}$	-16.3
$d_{ m Na_2O+K_2O}$	20.1

Note. "Anhydrous" case from Iacono-Marziano et al. (2012).

Consider some volume of magma with gas bubbles in it which contains a total number of moles γ_{tot} . The total moles is the sum of the moles of magma (γ_{magma}), and the moles of gas in bubbles suspended in that magma (γ_{gas}):

$$\gamma_{\text{tot}} = \gamma_{\text{gas}} + \gamma_{\text{magma}} \tag{A4}$$

Within this same volume of magma, the total moles of carbon ($\gamma_{\rm C}^{\rm tot}$) is equal to the moles of carbon in the gas phase ($\gamma_{\rm C}^{\rm gas}$) and the moles of carbon dissolved in the magma ($\gamma_{\rm C}^{\rm magma}$) combined.

$$\gamma_{\rm C}^{\rm tot} = \gamma_{\rm C}^{\rm gas} + \gamma_{\rm C}^{\rm magma} \tag{A5}$$

We assume that the only carbon bearing molecule that can dissolve in the magma is CO_2 , therefore $\gamma_C^{\text{magma}} = \gamma_{CO_2}^{\text{magma}}$. Dividing by γ_{tot} and expanding gives

$$\frac{\gamma_{\rm C}^{\rm tot}}{\gamma_{\rm tot}} = \frac{\gamma_{\rm gas}}{\gamma_{\rm tot}} \frac{\gamma_{\rm C}^{\rm gas}}{\gamma_{\rm gas}} + \frac{\gamma_{\rm magma}}{\gamma_{\rm tot}} \frac{\gamma_{\rm CO_2}^{\rm magma}}{\gamma_{\rm magma}}$$
(A6)

We can replace $\frac{\gamma_{\text{magma}}}{\gamma_{\text{tot}}}$ with $1 - \frac{\gamma_{\text{gas}}}{\gamma_{\text{tot}}}$ using Equation (A4). This leaves us with

$$\frac{\gamma_{\rm C}^{\rm tot}}{\gamma_{\rm tot}} = \frac{\gamma_{\rm gas}}{\gamma_{\rm tot}} \frac{\gamma_{\rm C}^{\rm gas}}{\gamma_{\rm gas}} + \left(1 - \frac{\gamma_{\rm gas}}{\gamma_{\rm tot}}\right) \frac{\gamma_{\rm CO_2}^{\rm magma}}{\gamma_{\rm magma}} \tag{A7}$$

Notice that $\frac{\gamma_{\text{CO}_2}^{\text{magma}}}{\gamma_{\text{magma}}}$ is just x_{CO_2} (the mol fraction of CO_2 in the magma). Also, we assume that CO_2 , CO and CH_4 are the only carbon-bearing gas species, so $\gamma_{\text{C}}^{\text{gas}} = \gamma_{\text{CO}_2}^{\text{gas}} + \gamma_{\text{CO}}^{\text{gas}} + \gamma_{\text{CH}_4}^{\text{gas}}$. Making substitutions gives

$$\frac{\gamma_{\rm C}^{\rm tot}}{\gamma_{\rm tot}} = \frac{\gamma_{\rm gas}}{\gamma_{\rm tot}} \frac{\gamma_{\rm CO_2}^{\rm gas} + \gamma_{\rm CO}^{\rm gas} + \gamma_{\rm CH_4}^{\rm gas}}{\gamma_{\rm gas}} + \left(1 - \frac{\gamma_{\rm gas}}{\gamma_{\rm tot}}\right) x_{\rm CO_2} \tag{A8}$$

Assuming the ideal gas law, $\gamma_i^{\rm gas}/\gamma_{\rm gas}=P_i/P$. Also, to make the equation more manageable, we substitute $\alpha_{\rm gas}=\frac{\gamma_{\rm gas}}{\gamma_{\rm tot}}$.

$$\frac{\gamma_{\rm C}^{\rm tot}}{\gamma_{\rm tot}} = \frac{P_{\rm CO_2} + P_{\rm CO} + P_{\rm CH_4}}{P} \alpha_{\rm gas} + (1 - \alpha_{\rm gas}) x_{\rm CO_2} \tag{A9}$$

Magma sometimes freezes deep in the Earth as a glass before it releases any volatiles. Researchers measure volatiles, like CO_2 , in such glasses and report concentrations in terms of mass fractions (Wallace et al. 2015). To stay consistent with these unit conventions, we indicate the total carbon in undegassed magma as a mass fraction of CO_2 ($m_{CO_2}^{tot}$). We can convert the mass fraction to a mole fraction using Equation (3):

$$\frac{m_{\text{CO2}}^{\text{tot}}}{x_m \mu_{\text{CO2}}} = x_{\text{CO2}}^{\text{tot}} = \frac{\gamma_{\text{CO2}}^{\text{tot}}}{\gamma_{\text{tot}}} = \frac{\gamma_{\text{C}}^{\text{tot}}}{\gamma_{\text{tot}}}$$
(A10)

Substituting Equation (A9) into Equation (A10) gives

$$\frac{m_{\text{CO}_2}^{\text{tot}}}{\mu_{\text{CO}_2} x_m} = \frac{P_{\text{CO}_2} + P_{\text{CO}} + P_{\text{CH}_4}}{P} \alpha_{\text{gas}} + (1 - \alpha_{\text{gas}}) x_{\text{CO}_2}$$
(A11)

Equation (A11) is identical to Equation (11).

A.3. Graphite saturation and the solubility of CO, CH₄, and H₂

Several studies have shown that degassing can be affected by graphite saturation of of magma (Hirschmann & Withers 2008) or by the solubility of CO, CH_4 , and H_2 in magma (Wetzel et al. 2013; Ardia et al. 2013; Hirschmann et al. 2012). Our model for outgassing speciation used throughout the main text does not account for these complications, although here we show that this assumption is valid and does not significantly change our results.

Consider the following equilibrium.

$$C + O_2 \leftrightarrow CO_2$$
 (A12)

$$K_9 = \frac{f_{\rm CO_2}}{a_{\rm C}f_{\rm O_2}} \approx \frac{P_{\rm CO_2}}{a_{\rm C}f_{\rm O_2}}$$
 (A13)

Here, K_9 is the equilibrium constant given by $\exp(47457/T + 0.136)$, and $a_{\rm C}$ is the activity of carbon. To incorporate graphite saturation into our model, we first calculate outgassing specation using the model described in the main text (Section 2.1). Next, we check for graphite saturate by claculating the activity of carbon using Equation (A13). If $a_{\rm C} < 1$, then we assume the melt is not gaphite saturated and that the calculation is valid. If $a_{\rm C} > 1$, then we assume graphite is saturated and recalculate outgassing speciation by replacing the carbon conservation equation (Equation (11)), with the graphite saturation equation with $a_{\rm C} = 1$ (Equations (A13)).

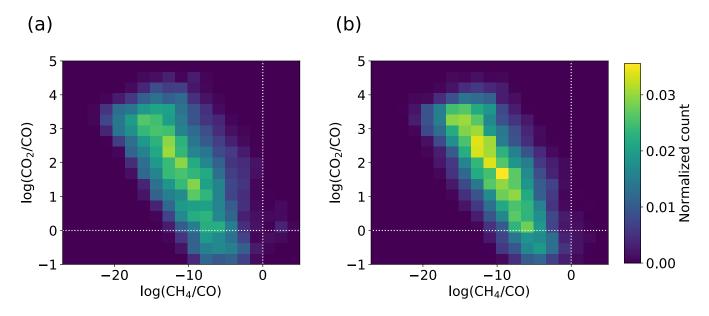


Figure 11. Identical to Figure 3, except here we account for graphite saturation in the melt. Like Figure 3 (a) is for ocean worlds and (b) is for Earth-like worlds. Graphite saturation has a small effect on the results.

Figure 11 is identical to Figure 3, except Figure 11 accounts for graphite saturation. Graphite saturation appears to have a small effect on the results, therefore it is justified to ignore it.

To incorporate the solubility of H_2 , CH_4 , and CO into our model we add the following solubility relationships to or system of original outgassing equations (Section 2.1).

$$\exp(-11.403 - 0.000076P) = K_5 = \frac{x_{\text{H}_2}}{f_{\text{H}_2}} \approx \frac{x_{\text{H}_2}}{P_{\text{H}_2}}$$
(A14)

$$\exp(-7.63 - 0.000193P) = K_6 = \frac{x_{\text{CH}_4}}{f_{\text{CH}_4}} \approx \frac{x_{\text{CH}_4}}{P_{\text{CH}_4}}$$
(A15)

$$\exp(-41.02 - 0.00056P) = K_7 = \frac{x_{\text{Fe(CO)}_5}}{a_{\text{Fe}} f_{\text{CO}}^5} \approx \frac{x_{\text{Fe(CO)}_5}}{a_{\text{Fe}} P_{\text{CO}}^5}$$
(A16)

Here, pressure dependent equilibrium constants K_5 , K_6 and K_7 are from Hirschmann et al. (2012), Ardia et al. (2013), and Wetzel et al. (2013), respectively. For Equation (A16), we take the activity of iron to be $a_{\rm Fe}=0.6$ based on the experiments in Wetzel et al. (2013). Also, we only include the Equation (A16) when the $f_{\rm O_2}<{\rm IW}$ -0.55 (IW is the Iron-Wustite mineral buffer) because Wetzel et al. (2013) only observed CO dissolved in magma for really low oxygen fugacity.

We also alter the carbon and hydrogen atom conservation equations in order to accommodate for new molecules in the melt.

$$\frac{m_{\text{CO}_2}^{\text{tot}}}{\mu_{\text{CO}_2} x_m} = \frac{P_{\text{CO}_2} + P_{\text{CO}} + P_{\text{CH}_4}}{P} \alpha_{\text{gas}} + (1 - \alpha_{\text{gas}}) \left(x_{\text{CO}_2} + x_{\text{CO}} + x_{\text{CH}_4} \right) \tag{A17}$$

$$\frac{m_{\rm H_2O}^{\rm tot}}{\mu_{\rm H_2O}x_m} = \frac{P_{\rm H_2O} + P_{\rm H_2} + 2P_{\rm CH_4}}{P}\alpha_{\rm gas} + (1 - \alpha_{\rm gas})(x_{\rm H_2O} + x_{\rm H_2} + 2x_{\rm CH_4})$$
(A18)

Here, x_i is the mol fraction of species i in the melt.

Figure 12 is identical to Figure 3, except 12 accounts for H₂, CH₄ and CO solubility in magma. The solubility of these three molecules has a small effect on the results, therefore they can be ignored.

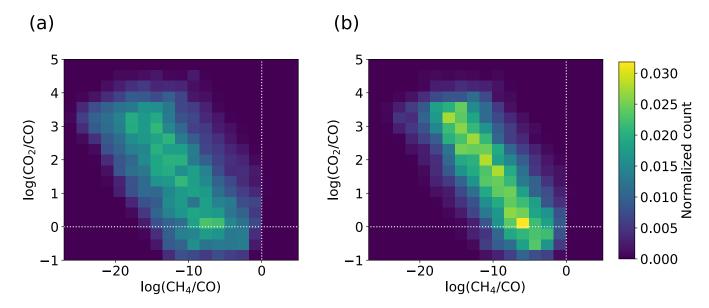


Figure 12. Identical to Figure 3, except here we account for the solubility of H₂, CH₄ and CO in the melt. Like Figure 3 (a) is for ocean worlds and (b) is for Earth-like worlds. H₂, CH₄ and CO solubility has a small effect on the results.

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