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SUPPLEMENTARY MATERIAL FOR:

Temporal variations of helium isotopes in volcanic gases quantify pre-eruptive refill and pressure buildup in magma reservoirs: The case of Mount Etna

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1. QUANTITATIVE MODELING

1.1 He isotope mixing

We model the He isotope mixing between the high-$^3He/^4He$ endmember outgassed from the central storage zone where the deep input enters, and the low-$^3He/^4He$ endmember coming from more distal magma batches virtually isolated from the innermost portion (Fig.2). In this Section, we will use R in place of $^3He/^4He$ to keep the notation simpler.

During periods far from a magmatic deep input, which we refer to as background periods (subscript "B"), the flow (Q_H) of the high-$^3He/^4He$ endmember sums to the flow (Q_L)of the low-$^3He/^4He$ endmember, so that the total gas flow toward surface is $Q_B=Q_L+Q_H$. A site S is fed by a fraction $Z_S=Q_{S,H}/Q_H$ of the high-R endmember and a fraction $Y_S=Q_{S,L}/Q_L$ of the low-R endmember, $Q_{S,L}$ and $Q_{S,H}$ being by definition the flows of two endmembers at the site. If we define $z_S=Z_S/(Z_S+Y_S)$ and $y_S=Y_S/(Z_S+Y_S)=(1-z_S)$, then the total flow at the site ($Q_{S,B}$) is:

$$Q_{S,B}=Q_{S,L}+Q_{S,H}=Z_SQ_H+Y_SQ_L=[z_SQ_H+(1-z_S)Q_L](Z_S+Y_S) \quad (1)$$

If $He_{S,B}$ and $R_{S,B}$ are He concentration and isotopic composition at site S, the mass balances for ^4He and ^3He are, respectively:

$$He_{S,B}Q_{S,B}=[z_SQ_HHe_H+(1-z_S)Q_LHe_L](Z_S+Y_S) \quad (2a)$$
$$R_{S,B}He_{S,B}Q_{S,B}=[z_SQ_HHe_HR_H+(1-z_S)Q_LHe_LR_L](Z_S+Y_S) \quad (2b)$$

where subscripts "L" and "H" refer to low-R and high-R endmember, respectively. We recall that all of the background parameters ($He_{S,B}$, $R_{S,B}$, $Q_{S,B}$ and Q_B) are associated to a rest periods of the volcano.

In contrast, during an unrest, a deep input enters the innermost zone (namely that more directly connected to the magma source) and causes overpressure in a part of this zone. The overpressurized volume, which we will refer to as the magma chamber, will release large amounts of volatiles which obviously have the He concentration and isotope ratio of the high-R endmember (He_H and R_H, respectively; see above). If $Q_{g,o}(t)$ indicates this time-dependent gas flow output from the chamber (subscripts "g" and "o" mean *gas* and *output*, respectively), this flow sums to the above mass balances. In analogy with Z_S, we define $X_S=Q_{S,g,o}(t)/Q_{g,o}(t)$ that is the fraction of $Q_{g,o}(t)$ reaching the site, and also $x_S=X_S/(Z_S+Y_S)$. Then, the equations 2a and b become, respectively:

$He_{S,T}Q_{S,T} = [x_S Q_{g,o}(t)He_H + z_S Q_H He_H + (1- z_S)Q_L He_L](Z_S + Y_S)$ (3a)

$R_{S,T}He_{S,T}Q_{S,T} = [x_S Q_{g,o}(t)He_H R_H + z_S Q_H He_H R_H + (1- z_S)Q_L He_L R_L](Z_S + Y_S)$ (3b)

where $He_{S,T}$ and $R_{S,T}$ are He concentration and isotope ratio at site S during the input ($R_{S,T}$, $He_{S,T}$ and $Q_{S,T}$ are in theory time-depending because function of $Q_{g,o}(t)$). Therefore, He isotope ratio at site can be compute by:

$$R_{S,T} = \frac{Q_{g,o}(t)He_H R_H x_S + Q_H He_H R_H z_S + Q_L He_L R_L (1 - z_S)}{Q_{g,o}(t)He_H x_S + Q_H He_H z_S + Q_L He_L (1 - z_S)} \quad (4)$$

As we will see in Parameterization (section 2.1), all the intensive quantities in equation 4 will be externally constrained while $Q_{g,o}(t)$ will be computed by the physical model of the magma chamber. In contrast, measurements of total volcanic gas flow at background can constrain the amount Q_B (see section 2.7), but not its components Q_H and Q_L (remind that Q_B=Q_L+Q_H). Therefore, by means of equations 2a,b, we firstly convert equation 4 to:

$$R_{S,T} = \frac{Q_{g,o}(t)He_H R_H x_S + Q_{S,B}He_{S,B}R_{S,B} /(Z_S + Y_S)}{Q_{g,o}(t)He_H x_S + Q_{S,B}He_{S,B} /(Z_S + Y_S)} \quad (5)$$

In the hypothesis that at background (volcano rest period) $Q_H \approx Q_L$ (see section 3 for effects of this assumption), it can be demonstrated that $Q_{S,B}/(Z_S + Y_S) = Q_B/2$. Thus equation 5 simplifies to:

$$R_{S,T} = \frac{Q_{g,o}(t)He_H R_H x_S + 0.5Q_B He_{S,B}R_{S,B}}{Q_{g,o}(t)He_H x_S + 0.5Q_B He_{S,B}} \quad (6)$$

If rename He_H, R_H and $R_{S,T}$ as He_o, R_o and $R_S(t)$, respectively, equation 6 converts to:

$$R_{S,T} = \frac{Q_{g,o}(t)He_o R_o x_S + 0.5Q_B He_{S,B}R_{S,B}}{Q_{g,o}(t)He_o x_o + 0.5Q_B He_{S,B}} \quad (7)$$

that is the mixing equation 1 reported in Methods.

1.2 The magma chamber

The magma chamber is a box-shaped reservoir having horizontal section A_{ch} and height H, located in an elastic medium (Fig.2). Input of melt plus gas bubbles into the chamber is allowed from the floor (e.g a fracture). We assume a well-mixed reservoir where the input plume can be dispersed in the body of melt, according to Woods and Cardoso (1997) and Phillips and Woods (2001). Owing to their rise speed, gas bubbles produce a gas flux generating a foam layer above the melt (which we will assume to consist of gas only). Gases in the foam layer can be lost from the roof of the chamber due to the permeability of the wall-rock. We also assume that an output of melt can occur by a conduit (open fractures) at a chamber flank. In such conditions, pressure-volume evolution of the reservoir is controlled by the volumetric flows from and into the chamber, coupled to the elastic deformation of wall-rock, melt-plus-bubbles mixture and foam layer. We do not include volume changes due to temperature variations and related exsolution-crystallization processes, as well as resorption of gas bubbles in melt, while discussing the rationale of this choice in Section 2.3.

In modeling the chamber deformation, we follow the approach by Woods and Huppert (2003), modified to take into account the effects of input and output of gases. This approach, although having no pretentiousness to be complete, has been displayed to capture the key processes ruling chamber overpressure. Following Woods and Huppert (2003), the pressure changes of the chamber over time can be described by the simplified relation:

$$f\frac{d\Delta P}{dt} = Q_i - Q_o \tag{8}$$

where ΔP is chamber overpressure, namely the pressure excess with respect to magmastatic one:

$$\Delta P = P_m - \rho_m g d \tag{9}$$

being d the depth of the chamber top, P_m and ρ_m pressure and density of magma in the chamber being at temperature T_m. As chamber walls are elastic, the overpressure will be equal in all the chamber. In equation 8, f is the chamber volume divided by the effective compressibility of the melt-gas-rock system; Q_i and Q_o are the volumetric flows into and from the chamber respectively. Here we consider that input flow from below consists of both melt ($Q_{m,i}$) and gas ($Q_{g,i}$) mixed together, thus the input term can be written as:

$$Q_i = Q_{g,i} + Q_{m,i} = c_{g,i}Q_i + (1-c_{g,i})Q_i \tag{10}$$

being $c_{g,i}$ the volume fraction of gas in the magmatic input from below. We recall that, if n_g is the mass fraction of free gas in the magma chamber, then the free gas volume fraction will be:

$$c_g = \frac{n_g/\rho_g}{n_g/\rho_g + (1-n_g)/\rho_m} \tag{11}$$

where ρ_g is the density of the gas phase. As concerns the output from chamber (Q_o in Eq.8), we assume that two types of mass loss can occur, namely i) flow of gas through the rocks of the roof ($Q_{g,o}$) and ii) flow of melt from a open fracture ($Q_{m,o}$) located at some height of the chamber side (but not in the foam layer). Therefore:

$$Q_o = Q_{g,o} + Q_{m,o} \tag{12}$$

where the two flow terms on right hand will be detailed later.
In equation 8, we can write f as sum of two terms (Woods and Huppert, 2003):

$$f = f_m + f_w \tag{13}$$

where f_m is volume-compressibility ratio of the magma, namely melt plus dispersed bubbles and gas foam layer, and f_w is the ratio between chamber volume divided by the wall-rock bulk modulus ($f_w = V_{ch}/\beta_w$). As concerns f_m we use the Woods and Huppert (2003)'s formulation that takes into account the fact that the magma is a mixture of melt and gas:

$$f_m = M_m\left[\frac{n_g}{\rho_g P}+\frac{1-n_g}{\rho_m\beta_m}\right] \tag{14}$$

where n_g is the mass fraction of gas in chamber (namely, gas in foam layer plus that in the dispersed bubbles); M_m is the mass of magma existing in the chamber volume; β_m is the bulk modulus of the melt. The term n_g strictly depends on the difference between input and output of gas in chamber:

$$\frac{dn_g}{dt}=\frac{\rho_g}{M_m}(Q_{g,i}-Q_{g,o}) \tag{15}$$

Because their floatation, bubbles can be lost from the melt, growing the foam layer at the chamber roof. At any time the gas amount n_g will be then sheared into dispersed bubbles in melt plus a fraction in the foam. If we call n_b the gas amount in the dispersed bubbles, it will evolve on time as found by Woods and Cardoso (1997):

$$\frac{dn_b}{dt}=\frac{Q_{g,i}\rho_g}{M_m}-\frac{v_b}{H}n_b \tag{16}$$

where v_b is the bubble rise speed, that can be derived from Stokes's law as soon as a bubble radius has been assumed. By definition, the difference between n_g and n_b at any time gives the mass fraction of gas in the foam layer, which can be converted into a foam thickness when taking into account the gas density and the roof area (A_{ch}).

We still need to define the gas and melt output terms in equation 12. According to Jaupart and Allegre (1991) and Kozono and Koyaguchi (2012), the gas flow through the rocks of an magma conduit can be modeled by Darcy law, and the permeability of near-field rocks controls the process. We assume a lithostatic pressure regime that, based on results of Corsaro and Pompilio (2004), closely matches the magmastatic gradient too. Finally, we assume that gas loss can only occur at roof whereas the side walls are considered as impermeable. Indeed, even considering a gas flux from side walls, it should be scaled by the bubble concentration in melt with respect to the gas flux from the foam at the roof, as the foam consists of gas only. Therefore the flux at walls would be by some orders of magnitude lower than at the roof. The Darcy flow from the chamber will be then driven by the vertical pressure gradient at the roof, being z the vertical axis (positive upwards):

$$Q_{g,o}=-\frac{k(\Delta P)\cdot A_{ch}}{\mu_g}\left(\frac{dP}{dz}\right)_{z=d} \tag{17}$$

where $k(\Delta P)$ denotes an overpressure-dependent permeability that will be discussed at Section 2.6, μ_g is gas viscosity, A_{ch} and d are the already mentioned roof area and depth, respectively. The gradient $(dP/dz)_{z=d}$ can be approximated to $-(P_m-P_h-\rho_g gh)/h$, where h is some characteristic vertical distance from chamber roof at which the pressure comes back to lithostatic value P_h (Jaupart and Allegre, 1991). Because $P_m-P_h = \Delta P+\rho_m gh$, equation 17 can be rewritten as:

$$Q_{g,o} = \frac{k(\Delta P)\cdot A_{ch}}{\mu_g}\left[\frac{\Delta P}{h}+(\rho_m-\rho_g)g\right] \tag{18}$$

On the other hand, the output of melt from the chamber is modeled as Poiseuille flow through a vertical conduit having circular section:

$$Q_{m,o} = \frac{\pi r^4}{8\mu_m Z}\Delta P \tag{19}$$

being μ_m the melt viscosity, r and Z the radius and height of the conduit respectively.

2. PARAMETERIZATION

2.1 Mixing parameters for He

The concentrations $He_{S,B}$ and He_o in equation 7 were constrained according to Paonita et al. (2012). The Authors in fact showed that the entire range of magmatic pressure feeding the investigated sites varies between 130 and 400 MPa. Depending on the type of degassing (open- or closed-system), the He content of the magmatic gases in this pressure range varies within a very narrow range (6 to 8 ppm). Therefore, to assume a single average value for both $He_{S,B}$ and He_o of 6 ppm is a very good approximation in equation 7. As concerns the isotope compositions, $R_{S,B}$ has been defined as the background value of the site *S*, so that it can be chosen as the He isotopic ratio measured at the site before the increase we are modeling. R_o can be constrained as equal or higher than the highest He isotope ratio measured in our dataset (i.d. the highest of P39), because the pressurized chamber outgases by definition the isotopic endmember at high-*R*. Following the results of Correale et al. (2014) based on fluid inclusion data, the value can be selected to 7.6 R/Ra.

2.2 The chamber: size, pressure and temperature

The peripheral sites take gases from a complex dike-and-sill structure located between 200 and 400 MPa (Paonita et al., 2012). This interval matches a pressure range of 5 to 12 km where De Gori et al. (2005) locate a magmatic conduit-like zone which would represent the main way of ascent for Etnean magmas. On this basis, we selected 300 MPa as the pressure of the chamber (meaning a d value around 8 km b.s.l.). The temperature of the resident Etnean basalt was considered to be 1150°C, close to the predicted liquidus of the melt (Corsaro et al., 2007). The volume of this reservoir is not well constrained. As recalled, seismic tomographies (Chiarabba et al., 2000; De Gori et al., 2005; Patanè et al., 2006) provide evidences of a conduit-like structure of magma rise at depth higher than 5 km b.s.l., overtopped by a wider and shallower zone at about 3 km b.s.l.. Ground deformation measurements show a well-defined and localized sources of overpressure in space and time, between 9 and 3 km (i.e., Bruno et al., 2012). Based on ground deformation data, an estimation of the pressurizing chamber gave volume around 3-4 km^3, centered at 8 km below the volcano summit (Bonaccorso et al., 2005), therefore we choose this size range as representative of the modeled chamber. As concerns the shape (i.e., it would have no role in Woods and Huppert (2003)'s approach that just deals with volume, nevertheless it affects the roof area available for outgassing and then the output flow in our model. More or less smoothed shapes (see Fig. 18 in Gudmundsson 2012) can be hardly defined, so we preferred to select a simple box-like geometry. For a chamber volume of 4 km^3 (see above), we assumed A_{ch} and H to be 4 km^2 and 1 km respectively. Variation of H/A_{ch} ratio will be discussed in Section 3.

2.3 Volatile content of melt

Based on the degassing path discussed by Paonita et al. (2012), Etnean magma at 300 MPa pressure of the chamber dissolves about 3.5 wt% of H_2O and 1800 ppm of CO_2. We consider these concentrations as being representative of the stored melt in the chamber. Similarly, a deep input reaching the chamber from depth and moving along the same degassing path enters the chamber by carrying such dissolved H_2O and CO_2 concentration, plus an amount of mixed H_2O-CO_2 vapor exsolved during its decompression. The composition of this vapor at equilibrium with the dissolved volatiles can be calculated to have ~65 mol% CO_2 (~35 mol% H_2O; Paonita et al., 2012), whereas the relative amount of gas phase with respect to the melt depends on initial conditions (initial volatile amount) and degassing mechanism (open, multistep, closed) which are not known. The magma chamber is therefore modeled to get inputs of both H_2O-CO_2-bearing melt and H_2O-CO_2 mixed gas with the selected compositions, whereas the ratio between gas and melt of the entering mass is discussed in Section 2.7. In the melt layer, gas is considered to be dispersed in bubbles. Average size of this bubbles is required for calculating v_b in equation 16 by Stock's law. Estimations of mean bubble diameter in Mount Etna products is available from Polacci et al. (2006).

Some complications could come from the exsolution-crystallization-resorption processes. The saturated magma can in fact exsolve gas by crystallization due to cooling. Cooling rates cannot be constrained well and can change by orders of magnitude (10^{-5} to10^{-9} K/s; Woods and Huppert, 2003), whereas the rate of crystallization on temperature can be derived from MELTS code (Ghiorso et al., 2002) simulation of Mount Etna basalt (0.002 kg of crystal / kg of melt × K from simulations by Correale et al., 2014). With an average cooling rate of 10^{-7} K/s, the melt would produce crystal fraction of <0.01 in one year (the time scale of our modeled changes), which would cause exsolution of almost 4×10^{-5} kg of H_2O-CO_2 vapor per kg melt (computed by using Papale et al., 2006). On the other hand, the deep magmatic input carries gases into the chamber while the growing magmatic overpressures (10-20 MPa) makes the stored melt slightly undersaturated. This could produce a resorption of some amount of volatiles. In detail, moving along the degassing path by Paonita et al. (2012), we calculate that about 10^{-4} kg of H_2O-CO_2 vapor per kg melt could re-dissolve at the maximum overpressure of 20 MPa. For both crystallization and resorption, we calculate that any change in the H_2O-CO_2 composition of the gas phase is extremely small (<1%), thus the gas composition can be assumed as a constant. The net result of the two competing processes is that more or less 5×10^{-5} kg of vapor per kg melt can be dissolved. Indeed this is a maximum value because bubbles will not really dispersed in all the melt, so resorption could occur at a lower extent. Anyway, this maximum amount has to be compared to the amount entering the chamber in order to appreciate the effect of neglecting crystallization-resorption. Mount Etna application deals with entering gas amounts of $\sim10^{-3}$ kg per kg of melt, therefore we are confident that to neglect these processes cannot change the main significance of our results.

2.4 Melt and gas densities and viscosities

Melt density and viscosity were computed for the Etnean basalt at the above conditions of P, T and dissolved gases by using MELTS code, and their values were assumed to be constant during the simulated evolution in chamber. Although changes could indeed occur due to crystallization, the low crystal fractions in magma we predicted above (<0.01), coupled to the modest density differences between crystal and melt, allows to consider the two properties as independent on changes of the crystal content.

Gas density was computed by the equation of state reported in Nuccio and Paonita (2001) at the chamber pressure. The gas was described as a H_2O-CO_2 mixture with the composition given in Sect. 2.3. Viscosity of CO_2 and steam at high temperature are similar and very poorly dependent on pressure. An average value of viscosity was adopted for the gas phase at 1150°C (Kaye & Laby Online, 2005).

2.5 Bulk modulus of melt, rock and gas

Three values of bulk modulus have to be set in the model, namely for wall-rock, melt and gas. In the case of wall-rock, this parameter is not well constrained as depending on temperature and stress state of material. Values for both Young's modulus and Poisson's ratio were estimated by Aloisi et al. (2002, 2011) along a depth profile below Mount Etna, and they were used to calculate a rock bulk modulus by standard equations for homogeneous isotropic materials.

The bulk modulus of the melt was computed by MELTS code for the Etnean basalt with the selected content of H_2O. Finally, bulk modulus of the gas was considered equal to pressure, as for an ideal gas (see Eq.14). The slight difference with respect to use of a compressibility coefficient derived from Nuccio and Paonita (2001) equation of state is negligible when compared to the order-of-magnitude difference with respect to the compressibility of both melt and rock.

2.6 Rock permeability

Permeability of wall-rock at the roof of the chamber is a key parameter. In conduit models with lateral gas loss this parameter has been considered as a constant, or to be dependent on the confining pressure and therefore on depth (Kozono et al., 2012).

In contrast, we have to consider that the growth of an overpressure in a magma chamber cause changes in the state of tensile stress in wall rocks. As a consequence we can hypothesize creation and opening of microfractures, and a related increase of rock permeability. The increase of permeability may be particularly large if fluid-driven fracturing processes are involved (Zencher et al., 2006). In this case, a more suitable physical quantity which influences the permeability within a rock would be the effective pressure, namely the difference between the pressure of fluids in microfractures with respect to the confining pressure (Morrow et al., 1986; Christensen and Ramananantoandro, 1988; Wu and Pruess, 2000; Faulkner, 2004). The dependence of permeability on fluid pressure can be very complex, due to the fractal geometry of fractures, crack opening and their propagation under internal fluid overpressure. Modeling of overpressure-dependent permeability in a fractured rock was performed by Zencher et al. (2006). They sketched a permeable rock as composed by a sequence of layers of intact porous rock, with a constant characteristic permeability $k°$, alternated with fractured layers. The latter ones consist of a series of parallel fractures having half-length l, relative distance D, and an opening being a function of the internal overpressure:

$$\Delta u = \pi(1-\nu) l \Delta P / G\Sigma \tag{20}$$

where G and ν are Young's modulus and Poisson's coefficient of the rock, and Σ is a coefficient computed by taking into account the interaction among dislocations (see Zencher et al., 2006). Overpressure would affect the permeability of the fractured layers according to (Zencher et al., 2006):

$$k(\Delta P)= \Delta u^3/12D - (\Delta u/D)k° + k° \tag{21}$$

In the limit of an fractured rock with no intact layers inside, equations 20-21 provide a proper relation between permeability and overpressure. In accordance with Jamtveit and Yardley (1997), we used a $k°$ value of 5×10^{-18} m^2, able to guarantee a lithostatic pressure gradient. By using Young's modulus and Poisson's coefficient from Sect. 5.4, the values of l and D where chosen to reproduce a ten-fold increase of permeability for overpressure close to 20 MPa, in the range discussed for a basalt rock by Zencher et al. (2006).

In our approximation of the Darcian flux from the chamber roof, the distance h in equation 18 can be regarded as the one at which the deviatoric stress due to the internal overpressure becomes negligible and it strictly depends on the shape of the chamber, presence of structures and conditions of overlying rock (Dragoni and Magnanensi, 1989; Gudmundsson, 2006; Simakin and Ghassemi, 2010; Bistacchi et al., 2012). In the simple case of a spherical chamber in an homogeneous elastic medium the deviatoric stress falls with the cube of the distance from the wall. For a chamber of 1000 m diameter the deviatoric stress would be decreased to 1/3 already 200 m far from the wall, suggesting that the rock layer closer to the roof would suffer the main effects of overpressure. Moreover, as permeability varies with the cube of the overpressure (see Eqs. 20-21), the permeability increase driven by overpressure would involve the rock portion still closer to the roof. In general, we can qualitatively guess this behaviour even considering that the decay function of the stress is different when changing the chamber shape. In real systems the framework could be also complicated by the conditions of wall rocks and, above all, in presence of faults and structures. Petrographic markers of strain in aureoles of igneous complexes suggest that the deviatoric stress decayed by ten folds when moving a few hundreds meters away from the igneous margin (Johnson et al., 2011). Based on the above considerations we selected h equals to 200 m as default value of our model for a chamber 1000 m high. We will show that this choice has not relevant impact on the results of the rest of the work (see Sect. 3).

2.7 Deep input in chamber and background flow

In the last forty years, Mount Etna has displayed a tendency to maintain an equilibrium between magma input and output (Wadge and Guest, 1981; Allard et al., 2006; Harris et al., 2011, 2012; Bonaccorso et al., 2013a), therefore the erupted volume can be considered a good proxy of the recharge volume. From January 2011 to April 2012 Mount Etna initiated a sequence of 25 lava fountains (Behncke et al., 2014), whose total emitted volume was estimated very well. The output of each fountain was in fact quantified by methods based on satellite and ground-based thermal images (Calvari et al., 2011; Ganci et al., 2012; Bonaccorso et al., 2013b), with values moving in the range of 1.8 to 3.0×10^6 m^3 (lava plus pyroclastics). Also, signals from deep borehole strainmeters recently installed in the western flank of the volcano provided an average erupted volume of 2.5×10^6 m^3 per each fountain (Bonaccorso et al., 2013b), with a total output of about 60×10^6 m^3 for the 25 episodes of 2010-2012. Topographic survey of the NSEC scoria cone and lava flows gave ~35×10^6 m^3 of dense rock (Behncke et al., 2014). Taking into account the uncertainties in these estimations (±50%), a total erupted volume ranging from $50 \pm 10 \times 10^6$ m^3 can be considered as representative of the 2010-2012 activity. The eruptive activity started while $^3He/^4He$ was increasing and terminated with the minimum of $^3He/^4He$, before a new recharge event (Fig. 3), therefore we can reasonably assume that the whole erupted volume was provided by the single recharge event marked by the $^3He/^4He$ variation of 2010-2012. By dividing the average erupted volume by the time elapsed from the start of the $^3He/^4He$ increase up to the onset of the decrease, we achieve an average flow rate of melt deep input of about 0.95 m^3/s.

As concerns the gas input, a time-averaged total volatile output (H_2O>CO_2>SO_2) from Etnean crater have been estimated to be around 20 kt/d (Aiuppa et al., 2008), which could be somewhat higher if considering degassing from flanks and into aquifers (by about 10-20%; D'Alessandro et al., 1998). Starting from measured 20 kt/d output, we can convert it into 0.4 m^3/s ($Q_{i,g}$) at the gas density calculated at depth, meaning a volume fraction of gas $c_{g,i}$ around 0.33 (or 0.5 gas/melt ratio). In terms of mass, we mean that the gas input is about 10% of the total melt plus gas deep input, which is much higher than the amount of gas that the magma can dissolve in the crustal reservoir. Such excess of volatiles with respect to melt had already been revealed at Mount Etna by Allard et al. (2006). In accordance with recent findings (Ferlito et al., 2014), it would involve flux of volatiles decoupled from melt which rise directly from the underlying mantle source.

Volatile output from the summit craters was also measured when Mount Etna was passively degassing and it gave a value around 7 kt/d (Aiuppa et al., 2008), resulting by a factor 3 to 5 lower than time-averaged degassing. The passive degassing value can be tentatively used to constrain the background gas flow Q_B to be inserted into equation 7, namely the mixed gas output coming from both high-$^3He/^4He$ and low-$^3He/^4He$ magmatic levels far from unrest periods marked by deep inputs. The same as done above, the passive degassing output is converted into a volumetric flow of 0.14 m^3/s.

2.8 The output conduit

Geometrical parameters are required to describe the output conduit of magma, namely radius r and vertical extension Z in equation 19. Indeed several dikes and fractures can drive magma output and, because any information is lacking on this aspect, we prefer to simplify the problem by modeling a single conduit. If assuming that this conduit would connect the top of the deep reservoir to the bottom of the shallower storage volume, we can estimate Z around 3 km. In contrast, we have no information on the radius r, thus it was constrained by considering that it is the key parameter in determining the highest value of overpressure reached during simulation. Our constraint for r was then that the highest overpressure computed by the simulation did not overcome the maximum value endurable by a magma chamber. Realistic values of overpressures are generally considered to be lower than 20 MPa, although the tensile strength of the wall-rocks depends on thermal state, depth, presence of cracks and regional stress field (Tait et al., 1989; Gudmundsson, 2012). Above this edge value, we can expect failure of bounding wall rocks and opening of new dikes. The formation of a dike nucleating from ~3 km b.s.l. and bypassing the whole shallow system was only evident in 2001 (Neri et al., 2005), and thus the selected 20-MPa threshold is suitable for estimating r during the 2010–2012 event. On the other hand, with the above selected rock permeability, overpressure below 15 MPa would cause that the maximum gas output from the chamber drops down to values three times as low as those derived from the measured plume flux during unrest periods (Section 2.7). Therefore, the value of r was chosen in order to constrain the maximum overpressure between 15 and 20 MPa. Moving the maximum value within this range, as well as shifting this permissible range toward higher or lower pressure, does not affect our results in a significant way (see Section 3).

3. MODEL SENSITIVITY

We numerically studied the effects of changing values of model parameters. We chiefly investigated how the changes affect the computed He isotope signal. Particularly, we focused on the time scale of the He isotope increase, namely the time to reach the highest and steady-state $^3He/^4He$ value of the simulated signal. From this point of view, the investigated parameters can be divided into two groups, based on the fact that they affect or not the time scale of the He isotope increase when moved within reasonable ranges of values. Changing values in parameters not affecting the time scale of $^3He/^4He$ growth modify the amplitude of the isotope variation (i.d., the difference between maximum $^3He/^4He$ value and background value), nevertheless the simulated signal can still reproduce the measured one by simply changing the value of the mixing fraction x_S, already considered as a fitting parameter in our approach. The key conclusion is that the selected values for these parameters have no major implication for our results. In contrast, parameters affecting the time scale of the $^3He/^4He$ increase can be modified so as to cause that the model is no more capable to fit the observed signal.

Parameters not affecting the time scale of the $^3He/^4He$ growth (and having scarce implication for our results) are shape of the box-like chamber (section and height), radius of the output conduit, gas/melt ratio of the input, distance from chamber where the effect of overpressure vanishes, overpressure-dependent permeability. Fortunately, this group includes the parameters affected by major uncertainty in our work.

Although the magma chamber volume was confidently constrained to 4 km^3, we were forced to assume a parallelepiped shape with a 4 km^2 horizontal section and 1 km height (A_{ch} and H, respectively). The area A_{ch} is that available for outgassing through the roof and then its change affects the output flow. Here we run the model by decreasing the roof area to 2 km^2 at a constant volume chamber. Such decrease lowers the output flow while consequently increasing the overpressure. The effect on He isotope signal is a more modest increment of the computed $^3He/^4He$ (Fig. S1). Very importantly, the time scale of the $^3He/^4He$ increase remains practically unchanged. This implies that the simulated $^3He/^4He$ signal can equally reproduce the measured one by simply changing the mixing fraction x_S (Fig. S1).

Similar outcome derives from exploring the effect of the parameter d, namely the distance from chamber where the effect of overpressure on rock permeability vanishes. Increasing d causes minor pressure gradients and then lower gas outputs. As a consequence, the $^3He/^4He$ increase is smaller (Fig. S1). The same as the roof area, the time scale of the $^3He/^4He$ increase remains practically unchanged, so that the measured $^3He/^4He$ signal can be reproduced by changing the x_S value (Fig. S1).

The radius of the output conduit affects dramatically the maximum overpressure reached during simulation. As stated, its value was selected in order to get maximum overpressure ranging between 15 and 20 MPa. Smaller radii will cause much higher overpressures while larger conduits would have opposite effects. In Figure S1 we show the $^3He/^4He$ signal achieved by decreasing the radius so as to increase the maximum overpressure from the value of 2010-2012 simulation (17.5 MPa) to the edge of 20 MPa. Although the amplitude of the $^3He/^4He$ increase changes consequently, the time scale remains practically the same (Fig. S1). It means that the observed ratios can be fitted by accordingly changing the mixing fraction x_S.

Further complication to be addressed is the weight of the value selected as tensile strength of wall-rocks, that could be different in relation to thermal and mechanical conditions (Sect. 2.8). Let we select a lower value of 10 MPa in place of 20 MPa. By using a larger conduit radius we will get maximum overpressure of simulation below the new permissible limit. As proven above, this type of change does not practically modify the time scale of the process, with no problem in fitting the $^3He/^4He$ signal by tuning the mixing fraction x_S.

On the other hand, parameters which affect the time scale of $^3He/^4He$ growth are magma input rate, chamber volume, rock and magma bulk moduli, all of them being acceptably constrained in our model. For instance, changes in magma input rate and chamber volume have opposite effects: an increase of magma input rate or a decrease in chamber volume determine faster pressurization, even if we use a larger conduit in order to maintain the maximum overpressure value within the given range. The gas output obviously increases in the case of higher input rates, while it decreases when considering a smaller chamber. As a consequence, the amplitude of the $^3He/^4He$ increment will result higher in the former case and lower in the latter one. Figure S1 displays the $^3He/^4He$ increase in the case of an input rate three times as high as that of 2010-2012 event, or a magma chamber about three times as small as the default value of 4 km^3. It is clear that 1) the time scale of the simulated signal becomes notably shorter and 2) the calculation is no more compatible with the observed isotope data. Anyhow we move the value of x_S we are not able to reproduce the observed isotopic variation.

Finally, we quantify the effects of the assumption $Q_H \approx Q_L$ in the mixing model. By using the relation $Q_B=Q_L+Q_H$ and $Q_{S,B}=[z_S Q_H + (1-z_S)Q_L](Z_S+Y_S)$ in section 1.1, we achieve:

$$Q_{S,B}/(Z_S+Y_S) = Q_L-2Q_L z_S+Q_B z_S \tag{22}$$

In the case $Q_H \approx Q_L$ equation 22 becomes $Q_{S,B}/(Z_S + Y_S) = Q_B/2$ as stated in calculating equation 6. To explore the weight of this constraint, we provide a numerical example. Let we suppose that $Q_H = 3Q_L$, thus equation 22 becomes:

$$Q_{S,B}/(Z_S + Y_S) = Q_B/4 + Q_B z_S/2 \tag{23}$$

The right member becomes $Q_B/2$ if z_S=1/2, the same as the case of $Q_H \approx Q_L$. The edge conditions of z_S=0 and z_S=1 give $Q_B/4$ and $3/4Q_B$. In the case of St site, use of $Q_B/4$ in place of $Q_B/2$ in equation 6 requires to increase x_S from 0.82 to 0.90 to achieve the same fit of the observed $^3He/^4He$ signal. From a general point of view, the $Q_H \approx Q_L$ constraint can only cause slightly different values of x_S in the fitting. Again, it cannot change the time scale of the process, therefore it does not affect the meaning of our results.

4. THE INVESTIGATED VENT SITES AT MOUNT ETNA

The four studied gas emissions are located at the base of Mount Etna and consist of gas coming out through water, mud or soil. The main gas species is CO_2 except for Fd where is CH_4. The sampling frequency ranged from twice a month during quiescence to twice a week during periods of volcanic unrest. In addition, fumaroles at the summit craters of the volcano have been sampled since 2007 and, given the site inaccessibility, only during the summer period, therefore they were not used for model application in this work.

Many authors investigated the spatial and temporal characteristics of these gases and their relation with volcanic activity (see references in Paonita et al., 2012). There is agreement that different magmatic levels feed the discharges as a function of their locations: the more the distance of the emissions from the volcano axis and the deeper the magma feeding them, with crater fumaroles fed from the shallowest magma portions.

The study of indicators of magma degassing (He/Ar, Ar/CO_2, C isotopes) allowed Paonita et al. (2012) to calculate pressures(depths) of the magmatic zones which feed the gas discharges. The peripheral vents, having He/Ar between 0.5 and 3.5, are mainly fed by magma that degases in the range 200-400 MPa, whereas the crater fumaroles (He/Ar from 2 to 5) get also gas contributions from exsolution at lower pressure-depth (up to 130 MPa). At the peripheral sites, shallow interaction of gas with aquifers modifies their geochemistry, so that the original magmatic signature needs to restored by using models of gas-aquifer interaction (Caracausi et al., 2003). This secondary process has no detectable effects on He isotope ratios of the vents, if excluding the addition of trivial amounts of air (Caracausi et al., 2003). Also, the process does not occur in crater fumaroles, where He/Ar vs. $\delta^{13}C_{CO2}$ show that, rather than simple decompression-driven degassing, exsolution of magmatic gases at high depth followed by mixing with gases exsolved at shallower magmatic levels is the most likely scenario. The degassing-mixing process accounts for an efficient bubble-melt decoupling, pointing to a system composed of horizontally elongated and dike-like structures that inhibit magma ascent.

REFERENCES CITED

Aiuppa A., Giudice G., Gurrieri S., Liuzzo M., Burton M., Caltabiano T., McGonigle A.J.S., Salerno G., Shinohara H., Valenza M. (2008) Total volatile flux from Mount Etna. *Geophys. Res. Lett.*, 35, L24302, doi:10.1029/2008GL035871.

Allard P., Behncke B., D'Amico S., Neri M., Gambino S. (2006) Mount Etna 1993–2005: Anatomy of an evolving eruptive cycle, Earth Sci. Rev., 78, 85-114, doi: 10.1016/j.earscirev.2006.04.002.

Aloisi, M., O. Cocina, G. Neri, B. Orecchio, and E. Privitera (2002), Seismic tomography of the crust underneath the Etna volcano, Sicily. *Phys. Earth Planet. Inter*. 134, 139-155.

Aloisi M., Mattia M., Monaco C., Pulvirenti F. (2011) Magma, faults, and gravitational loading at Mount Etna: The 2002–2003 eruptive period. *J. Geophys. Res*., 116, B05203, doi:10.1029/2010JB007909.

Behncke B., Branca S., Corsaro R.A., De Beni E., Miraglia L., Proietti C. (2014) The 2011–2012 summit activity of Mount Etna: Birth, growth and products of the new SE crater. J. Volcanol. Geotherm. Res. 270, 10-21.

Bistacchi A., Tibaldi A., Pasquarè F.A., Rust D. (2012) The association of cone–sheets and radial dykes: Data from the Isle of Skye (UK), numerical modelling, and implications for shallow magma chambers. Earth Planet, Sci. Lett. 339-340, 46-56.

Bonaccorso A., Cianetti S., Giunchi C., Trasatti E., Bonafede M., Boschi E. (2005) Analytical and 3-D numerical modelling of Mt. Etna (Italy) volcano inflation. *Geophys. J. Int.*, 163, 852-862.

Bonaccorso A., Calvari S. (2013a) Major effusive eruptions and recent lava fountains: Balance between expected and erupted magma volumes at Etna volcano. Geophys. Res. Lett. 40, 1-5, doi:10.1002/2013GL058291.

Bonaccorso A., Calvari S., Currenti G., Del Negro C., Ganci G., Linde A., Napoli R., Sacks S., Sicali A. (2013b) From source to surface: Dynamics of Etna's lava fountains investigated by continuous strain, magnetic, ground and satellite thermal data, Bull. Volcanol., 75, 690, doi:10.1007/s00445-013-0690-9.

Bruno V., Mattia M., Aloisi M., Palano M., Cannavò F., Holt W.E. (2012) Ground deformations and volcanic processes as imaged by CGPS data at Mt. Etna (Italy) between 2003 and 2008, *J. Geophys. Res.*, 117(B07208) doi:10.1029/2011JB009114.

Calvari S., Salerno G., Spampinato L., Gouhier M., La Spina A., Pecora E., Harris A.J.L., Labazuy P., Biale E., Boschi E. (2011) An unloading foam model to constrain Etna's 11–13 January 2011 lava fountaining episode. *J. Geophys. Res.*, 116, B11207, doi:10.1029/2011JB008407.

Caracausi A., Italiano F., Nuccio P.M., Paonita A., Rizzo A. (2003) Evidence of deep magma degassing and ascent by geochemistry of peripheral gas emissions at Mt. Etna (Italy): assessment of the magmatic reservoir pressure. *J. Geophys. Res.* 108, 2463-2484.

Chiarabba C., Amato A., Boschi E., Barberi F. (2000) Recent seismicity and tomographic modeling of the Mount Etna plumbing system. J. Geophys. Res., 105, 10923–10938.

Christensen N.I., Ramananantoandro, R. (1988) Permeability of the oceanic crust based on experimental studies of basalt permeability at elevated pressures. *Tectonophysics*, 149, 181–186.

Correale A., Paonita A., Martelli M., Rizzo A.L., Rotolo S., Corsaro R.A, Di Renzo V. (2014) A two-component mantle source feeding Mt. Etna magmatism: insights from the geochemistry of primitive magmas. *Lithos*,184-187C, 243-258.

Corsaro, R. A. and Pompilio M. (2004) Buoyancy-controlled eruption of magmas at Mt Etna. *Terra Nova*, **16**(1), 16 –22, doi:10.1046/j.1365-3121.2003.00520.x.

Corsaro, R.A., Miraglia L. and Pompilio, M. (2007) Petrologic evidence of a complex plumbing system feeding the July–August 2001 eruption of Mt. Etna, Sicily, Italy. *Bull. Volcanol.* **69**, 401–421.

D'Alessandro W., De Gregorio S., Dongarrà G., Gurrieri S., Parello F. and Parisi B. (1997) Chemical and isotopic characterization of the gases of Mount Etna (Italy). J. *Volcanol. Geotherm. Res.*, **78**, 65–76.

De Gori P., Chiarabba C. and Patanè D. (2005) Qp structure of Mount Etna: Constraints for the physics of the plumbing system. *J. Geophys. Res.*, **110**, B05303, doi:10.1029/2003JB002875.

Dragoni M., Magnanensi C. (1989) Displacement and stress produced by a pressurized, spherical magma chamber, surrounded by a viscoelastic shell. *Phys. Earth Planet. Inter*. 56, 316-328.

Faulkner D.R. (2004) A model for the variation in permeability of clay bearing fault gouge with depth in the brittle crust *Geophys. Res. Lett.* 31, L19611, doi:10.1029/2004GL020736.

Ferlito C., Coltorti M., Lanzafame G., Giacomini P. (2014) The volatile flushing triggers eruptions at open conduit volcanoes: Evidence from Mount Etna volcano (Italy). Lithos 184-187, 447-455.

Ganci G., Harris A.J.L., Del Negro C., Guehenneux Y., Cappello A., Labazuy P., Calvari S., Gouhier M. (2012) A year of lava fountaining at Etna: volumes from SEVIRI. *Geophys. Res. Lett*. 39, L06305, http://dx.doi.org/10.1029/2012GL051026.

Ghiorso M.S., Hirschmann M.M., Reiners P.W., Kress V.C. III (2002) The pMELTS: An revision of MELTS aimed at improving calculation of phase relations and major element partitioning involved in partial melting of the mantle at pressures up to 3 GPa. *Geochem. Geophys. Geosyst.* 3(5), 10.1029/2001GC000217.

Gudmundsson A. (2006) How local stresses control magma-chamber ruptures, dyke injections, and eruptions in composite volcanoes. *Earth Sci. Rev*. 79, 1-31.

Gudmundsson A. (2012) Magma chambers: Formation, local stresses, excess pressures, and compartments. *J. Volcanol. Geotherm. Res*., 237-238 (2012) 19–41.

Harris A.J.L., Steffke A., Calvari S., Spampinato L. (2011) Thirty years of satellite-derived lava discharge rates at Etna: Implications for steady volumetric output. *J. Geophys. Res*., 116, B08204, doi:10.1029/2011JB008237.

Harris A.J.L., Steffke A., Calvari S., Spampinato L. (2012) Correctionto "Thirty years of satellite-derived lava discharge rates at Etna: Implications for steady volumetric output". *J. Geophys. Res.*, 117, B08207, doi:10.1029/2012JB009431.

Kaye & Laby Online (2005) Tables of Physical & Chemical Constants (16th edition 1995), Version 1.0, www.kayelaby.npl.co.uk.

Kozono T., Koyaguchi T. (2012) Effects of gas escape and crystallization on the complexity of conduit flow dynamics during lava dome eruptions. J. Geophys. Res., 117(B08204), doi:10.1029/2012JB009343.

Jamtveit B., Yardley B. (1997) Fluid Flow and Transport in Rocks: Mechanisms and effects. Chapman & Hall, London, UK, pp. 315.

Jaupart, C., Allegre C. J. (1991) Gas content, eruption rate and instabilities of eruption regime in silicic volcanoes. *Earth Planet. Sci. Lett.*, 102(3-4), 413-429, doi:10.1016/0012-821X(91)90032-D.

Johnson S.E., Jin Z.-H., Naus-Thijssen F.M.J., Koons P.O. (2011) Coupled deformation and metamorphism in the roof of a tabular midcrustal igneous complex. *GSA Bulletin* 123 (5/6) 1016-1032, doi: 10.1130/B30269.1.

Morrow, C.A., Bo-Chong, Z. & Byerlee, J.D., 1986. Effective pressure law for permeability of Westerly granite under cycling loading. *J. Geophys. Res*., 91, 3870-3876.

Nuccio P.M., Paonita A. (2001) Magmatic degassing of multicomponent vapors and assessment of magma depth: application to Vulcano Island (Italy), *Earth Planet. Sci. Lett.*, 193(3-4), 467-481.

Paonita A., Caracausi A., Iacono-Marziano G., Martelli M., A. Rizzo (2012) Geochemical evidence for mixing between fluids exsolved at different depths in the magmatic system of Mt Etna (Italy), Geochim. Cosmochim. Acta, 84, 380–394.

Papale P., Moretti R. and D. Barbato (2006) The compositional dependence of the saturation surface of H_2O + CO_2 fluids in silicate melts. *Chem. Geol.*, 229,78-95.

Patanè D., Barberi G., Cocina O., De Gori P., Chiarabba C. (2006) Time-resolved seismic tomography detects magma intrusions at Mount Etna, *Science*, 313, 821, doi:10.1126/science.1127724.

Phillips J.C., Woods A.W. (2001) Bubble plumes generated during recharge of basaltic magma reservoirs. *Earth Planet. Sci. Lett*., 186, 297-309.

Polacci M., Corsaro R.A., Andronico D. (2006) Coupled textural and compositional characterization of basaltic scoria: insights into the transition from Strombolian to fire fountain activity at Mount Etna, Italy. *Geology* 34, 201–204.

Simakin A.G., Ghassemi A. (2010) The role of magma chamber-fault interaction in caldera forming eruptions. *Bull. Volcanol*. 72, 85-101.

Tait S., Jaupart C., Vergniolle S. (1989) Pressure, gas content and eruption periodicity of a shallow crystallising magma chamber. *Earth Planet. Sci. Lett*. 92, 107-123,

Wadge G., Guest J.E. (1981) Steady-state magma discharge at Etna 1971–81. *Nature*, 294, 548–550, doi:10.1038/294548a0.

Woods A.W., Cardoso S.S.S. (1997) Bubble magma separation as a trigger of basaltic volcanic eruptions. *Nature*, 385, 518-520.

Woods A.W., Huppert H.E. (2003) On magma chamber evolution during slow effusive eruptions. J. Geophys. Res., 108(B8), 2403, doi:10.1029/2002JB002019.

Wu Yu-Shu, Pruess K. (2000) Integral solutions for transient fluid flow through a porous medium with pressure-dependent permeability. *Int. J. Rock Mech. Min. Sci*. 37, 51-61.

Zencher F., Bonafede M., Stefansson R. (2006) Near-lithostatic pore pressure at seismogenic depths: a thermoporoelastic model. *Geophys. J. Int.* 166, 1318–1334.

Figure DR1. Simulated He isotope signal (thick curve) fitting the 2010-2012 variation observed at St site (noisy curve) and the effects of changing the values of several parameters. The solid curves have been computed by changing the value of the indicated parameter while leaving unmodified the mixing fraction xS, as estimated for 2010-2012. The dashed curves have been re-fitted to the measured 3He/4He signal by changing the xS value.