

The effect of lunar declination on CO₂ degassing from central Italian Apennines

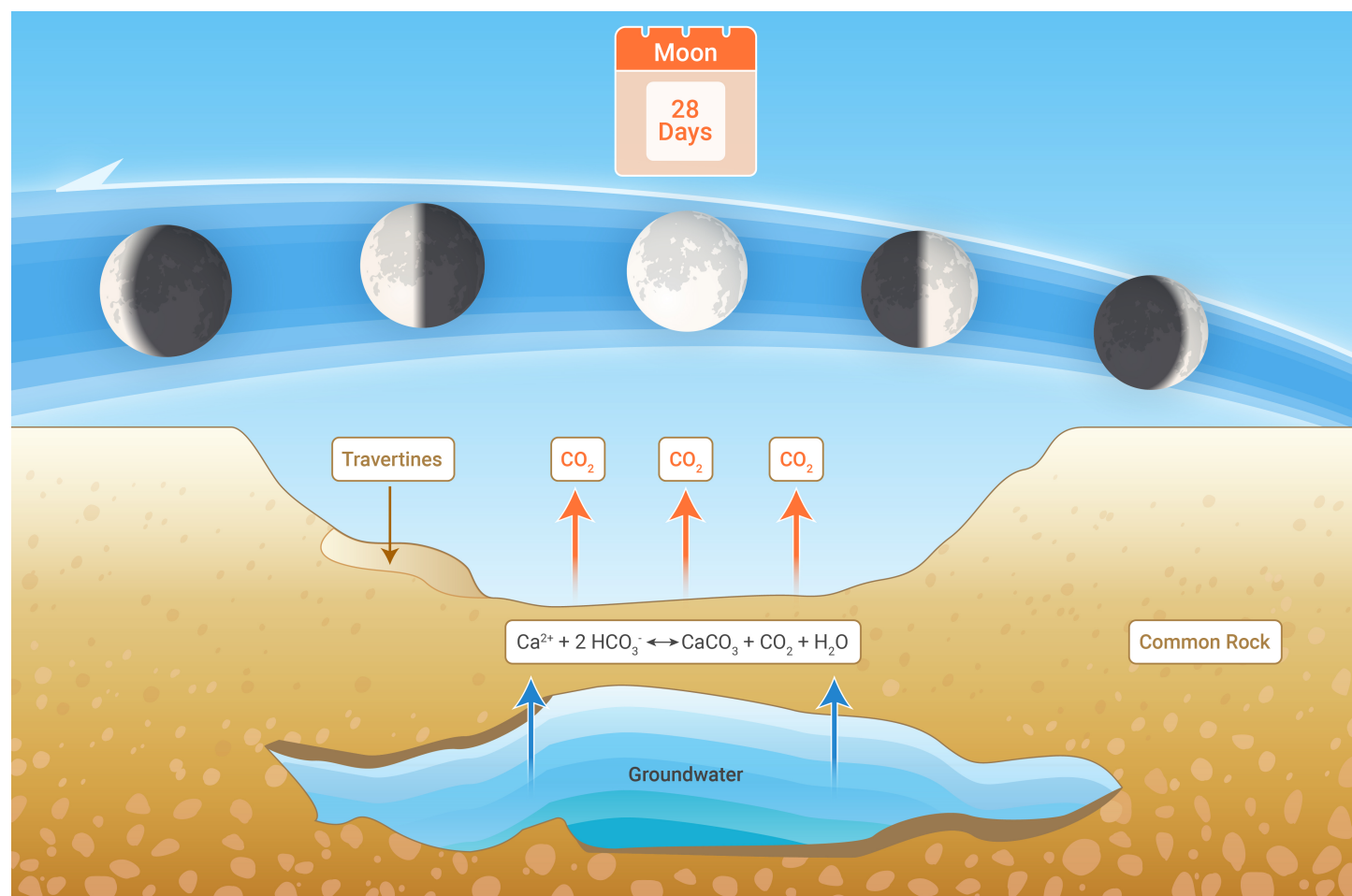
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GRAPHICAL ABSTRACT



PUBLIC SUMMARY

- The variation of aquifer's chemical composition is related on periodical lunar tides in central Italian Apennines.
- Lunar tides trigger fluid movement leading to periodic CO₂ degassing from over-pressured reservoirs.

The effect of lunar declination on CO₂ degassing from central Italian Apennines

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The periodical degassing from CO₂ over-pressured reservoirs may have serious consequences for the environment making urgent understanding the processes and forecasting the frequency. Prediction though needs methods that depends from temporal and spatial properties of hydro-chemical and physical reservoir characteristics that unfortunately are often lacking. We have analyzed surface emissions of CO₂ attributed to over-pressured CO₂-rich reservoirs in the Central Italian Apennines a zone characterized by significant periodical CO₂ degassing. Here aquifers are hosted in Mesozoic limestone with high pCO₂ groundwater and travertine deposits. We analyzed a 10-year temporal series and found that in the Apennines CO₂ flux and aquifer fluid composition are correlated with the lunar tides. In particular, our study reveals that low CO₂ flux corresponds with low lunar tidal potential values. We found a similar trend for dissolved calcium and water alkalinity, while pH values display a linear correlation with tidal cycles. The forces associated with tidal potentials are not capable of fracturing rock. However, they can, under certain conditions, drive the flow of fluids in over-pressured reservoirs, triggering sub-surface fluid movements that in turn modify the water-rock reactivity. In the central Apennines, we show that these movements result in increased dolomite dissolution and an eventual return to calcite equilibrium. In this case, dolomite dissolution breaks the rock releasing calcium into ground water, which leads to calcite equilibrium and in turn to the formation of significant quantities of travertine and the concomitant release of CO₂ in the atmosphere.

INTRODUCTION

The response of aquifers to earth and lunar tide strains has long been observed and recognized in several situations for more than one century^{1,2} and recently used to provide specific estimation storage and permeability of the aquifers.³ The potential impact of these strains has also been proposed as responsible for changes in stress and pore pressure in porous and in water-saturated formations.⁴ However, despite lunar tides are ubiquitous on the earth, the potential impacts of tides have never been evaluated in over pressured reservoirs. These reservoirs may occur naturally in particular geological situations or in low carbon strategies such as the geological sequestration of CO₂ or in the more traditional CO₂-enhanced recovering when CO₂ is injected into oil field to increase energy production.⁵ The distribution of CO₂ emission, in the Central Italian Apennines corresponds the formation of reservoirs fed by fluids rising through processes that imply the production and accumulation of CO₂ in the Earth's upper crust.^{6,7} Similarly, the geological CO₂ storage, a critical technology for environmental energy, produce oversaturated fluids conditions that may generated a CO₂ plume in the subsurface.⁸ The mechanisms that determine this process have complex and nonlinear interactions with geological properties (permeability, depth, thickness) and have been tentatively explained by the interaction between CO₂-saturated and CO₂-oversaturated aquifers.^{9,10} It is today difficult to predict the frequency of CO₂ variations using hydrogeological models because aquifer models assume that conditions are 'perfectly confined' or 'purely unconfined', whereas boundary conditions vary for regional-scale aquifers.^{3,11-14} Real temporal chemical measurements directly reflect these variations¹⁵ as they take into account possible low tectonic stress and periodical lunar tide oscillations that locally reshape the aquifers.^{11,16-24}

In our study, we have used the chemical composition records on springs and well in the Central Apennine area (Central Italy) collected over a 10-year period (2009-2018).¹⁰ We have calculated the CO₂ emissions in this area

confirming earlier estimates that indicate that the quantity of CO₂ released into the atmosphere from 2009-2018 in this zone is equivalent to the total amount of CO₂ released by Mount Etna (Italy), the Earth's premier CO₂-producing volcano over the same period. This amount is by over 1800 kt of CO₂ equivalent to a CO₂ flux of 10¹¹ mol yr⁻¹ that should correspond to 10% of the overall Earth flux for this range of time.^{18,25} In this paper we report a relation between lunar tides and the observed variation of CO₂ degassing during 10 years of monitoring in a non-volcanic area. We show for the first time the influence of lunar tides on water-rock reactivity and offer a geochemical mechanism explaining the observed variability of CO₂ degassing from the natural aquifers of central Apennines.

MATERIALS AND METHODS

The Italian Apennine region is home to several aquifers hosted in Mesozoic limestone and has frequent and significant gas emissions and numerous soda springs and wells with high pCO₂ groundwater and travertine deposits at the surface. Crustal thickness in the central Apennines is approximately 35 km, heat flow is normal and gravity anomalies are negative. The current tectonic deformation field is characterized by an extension in the axial zone of the Apennine chain and by a contraction responsible for periodic earthquakes of moderate magnitude (between 4 and 6), originating mainly in the upper crust.²⁶ They are generated by active lithospheric thrusts beneath the Apennine watershed and are generally associated with low-angle normal faults accommodating uplift of basement culminations. The large amount of CO₂ released into the atmosphere is most probably controlled by diffusive processes and the regional hydrogeological features of the degassing area (Figure 1). The zone under scrutiny covers 700 km² and is also characterized by numerous natural springs and wells that release CO₂ oversaturated water at the rate of hundreds of liters per second. The water releases CO₂ into the atmosphere and produces large travertine deposits that have been quarried since ancient roman times, over 2000 years ago. Field data used in this study (major and minor chemical fluid composition and dissolved CO₂) was collected from the 36 main springs and wells of the two large reservoirs, sampled between April 2009 and December 2018.¹⁰

We have recalculated pCO₂ partial pressure using field alkalinity and pH data and the overall water composition, using the PHREEQC software and thermodynamic database.²⁷ Springs, gas vents and travertine are potentially controlled by regional hydrology and meteoric water. The hydrological recharge is located in the Mesozoic limestone layers that overlay a thick and relatively impermeable cover of Oligocene-Neogene clastic sediments. Spring distribution is dictated by water circulation in the Mesozoic limestone, where water chemical composition is between Ca-SO₄ and Ca(Mg)-HCO₃, with very limited amounts of Na-HCO₃, Na-Cl and Na-SO₄.

To estimate the Lunar tides, we have considered that the Earth is spherically symmetrical, nonrotating and has self-gravity with elastic and isotropic properties in an initial equilibrium state. The Moon and the Sun are considered celestial bodies that produce a tidal effect on the Earth. Assuming M = Earth masse and m = Moon masse, every point P at the Earth's surface is subject to a force γ equal to:

$$\gamma = GMmPA'(PA')^3 \quad (1)$$

Where $PA'(PA')^3$ is the vector distance between the point P at the Earth's surface and the Moon's center of gravity A' , while G is the gravitational constant of Newton. The Earth is subject to an attraction force, γ_E , from the Moon equal to:

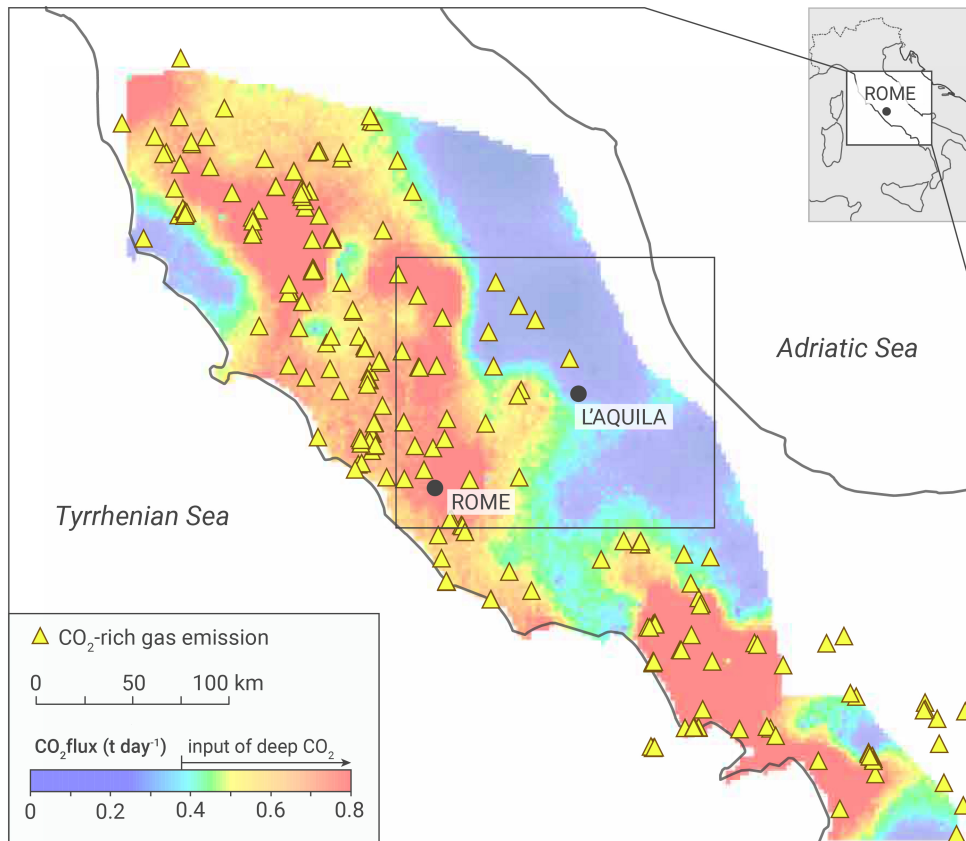


Figure 1. Map of CO₂ degassing and location of the investigated aquifers (from Chiodini et al. 2020).

Earth calculated by Eqn. 5 is reported in Figure 2. The lunar tidal signal (potential) is a continuous function due to the lunar attraction while data points concerning the pCO₂ partial pressure are independent on the lunar signal and should be randomly distributed along the continuous curve. We found that different pCO₂ partial pressure are systematically (95% of points) found at extrema of the lunar potential indicating that there is a very strong (at least 95%) correlation (the tidal signal being complex sinusoid) between concentration variations and the w_2 attraction. We were surprised by the existence of such a strong correspondence correlation, given that there is no clear mechanistic relationship between pCO₂ partial pressure and tidal potentials.

We have focused our attention on the data set gathered from June 2009-June 2010 because it was the largest and densest of the 10-year period. Figure 3A illustrates those variations in different

calcium concentrations are systematically (95% of values) found at extrema of the lunar potential. Given that calcium concentrations are not distributed along the continuous curve, it is

not possible to identify a continuous function between tidal potential variations and calcium concentrations using one-year data set. We can safely say however, based on our data sets, that minimum calcium concentrations are indeed related to weak Lunar tide potentials while higher calcium values are related to high Lunar potential values.

Figure 3B shows that variations in alkalinity concentration are concentrations are systematically (95% of points) at extrema of the lunar potential and not y distributed along the continuous curve. Similar to calcium, the lack of intermediate concentration values makes not possible identifying a continuous function for alkalinity. Figure 3C shows that variation of pCO₂ partial pressure that range between 0.01 atm. and 1.7 atm. is systematically found at the extrema of the lunar potential. Low pCO₂ (from 0.01 to 0.2 atm) corresponds to a weak Lunar potential while higher pCO₂ values (from 1.4 to 1.6 atm) correspond to stronger Lunar potential. These observations suggest that CO₂ emissions in the zone may be related to Lunar tidal forces in a discontinuous and apparently non-linear relationship.

We considered that it is possible that pCO₂ variations may reflect changes in chemical reactivity in the aquifer and if that is indeed the case, it should be reflected in fluid pH variations. In fact, variations in fluid pH globally reflect the complex process of water-rock interaction because pH participates in both carbonate and silicate mineral dissolution and precipitation reactions. Figure 4 illustrates that pH significantly varies between a minimum of 6.2 and a maximum of 8.2. However, unlike calcium, alkalinity and pCO₂ partial pressure, pH values are distributed along the lunar potential curve during the 27.55-day period indicates the presence of possible water-rock interaction processes responsible for the observed release of CO₂.

To explain the different behavior of pH, we have estimated the mineral saturation indexes (SI) of the water and found that sampled fluids are close to equilibrium with respect to calcite given that SI_{calcite} is between -0.7 and +0.3 with mean value of -0.1. It is likely that the principal process that controls groundwater composition is calcite dissolution in near-equilibrium conditions.¹⁸ The collected ground waters are under-saturated with respect to gypsum or anhydrite (CaSO₄, 2H₂O, CaSO₄ respectively) although it is possible that gypsum or anhydrite dissolution does take place in springs with high SO₄ content. When gypsum or anhydrite dissolves, it can facilitate dolomite dissolution because of the common ion effect.²⁹ This in turn could allow for

$$Y_E = \frac{GmAA'}{(AA')^3} \quad (2)$$

Where A is the Earth's center of gravity, AA' is the vector distance between the Earth's center of gravity and the Moon's center of gravity. The difference between the Moon's attraction on P and the Earth's attraction on the Moon, also called Earth tide force, $\mathbf{f}$, is the difference between $\mathbf{Y}$ and $\mathbf{Y}_E$. The gravitational force $\mathbf{f}$ can be expressed as a function of its potential w :

$$w = Gm \left[\frac{1}{PA} - \frac{-AP \cdot AA'}{(AA')^3} \right] \quad (3)$$

Since the position of point P on the Earth's surface is a function of the latitude z , we have expressed the Earth's tide potential w using Legendre polynomials:

$$w = \frac{Gm}{D} \times \sum_{n=0}^{\infty} \left(\frac{R}{D} \right)^n \times P_n(\cos z) \quad (4)$$

Where D is the distance between the Earth's center of gravity and the Moon's center of gravity, R is the distance between the given point P and the Earth's center of gravity and P^n is the Legendre polynomial function. The Earth's tidal potential w can be approximated to the second order Legendre polynomial term:

$$w_2 = \frac{GmR^2(3\cos^2 z - 1)}{2D^3} \quad (5)$$

Using lunar ephemeris and effective latitude positions were provided by the *Institut de Mécanique Céleste et de Calcul des Éphémérides* (IMCCE). We have calculated the attraction force generated by the Moon on the Earth (w), also called lunar tidal potential or tidal oscillations, using an average latitude of 43°. ²⁸

RESULTS

The principal lunar tide oscillations estimated by our mechanical calculations show a periodicity of 27.555 days. This value of periodicity results from the variation in the distance between the Earth and the Moon in the central Apennines. The lunar tidal potential responsible for the attraction force on the

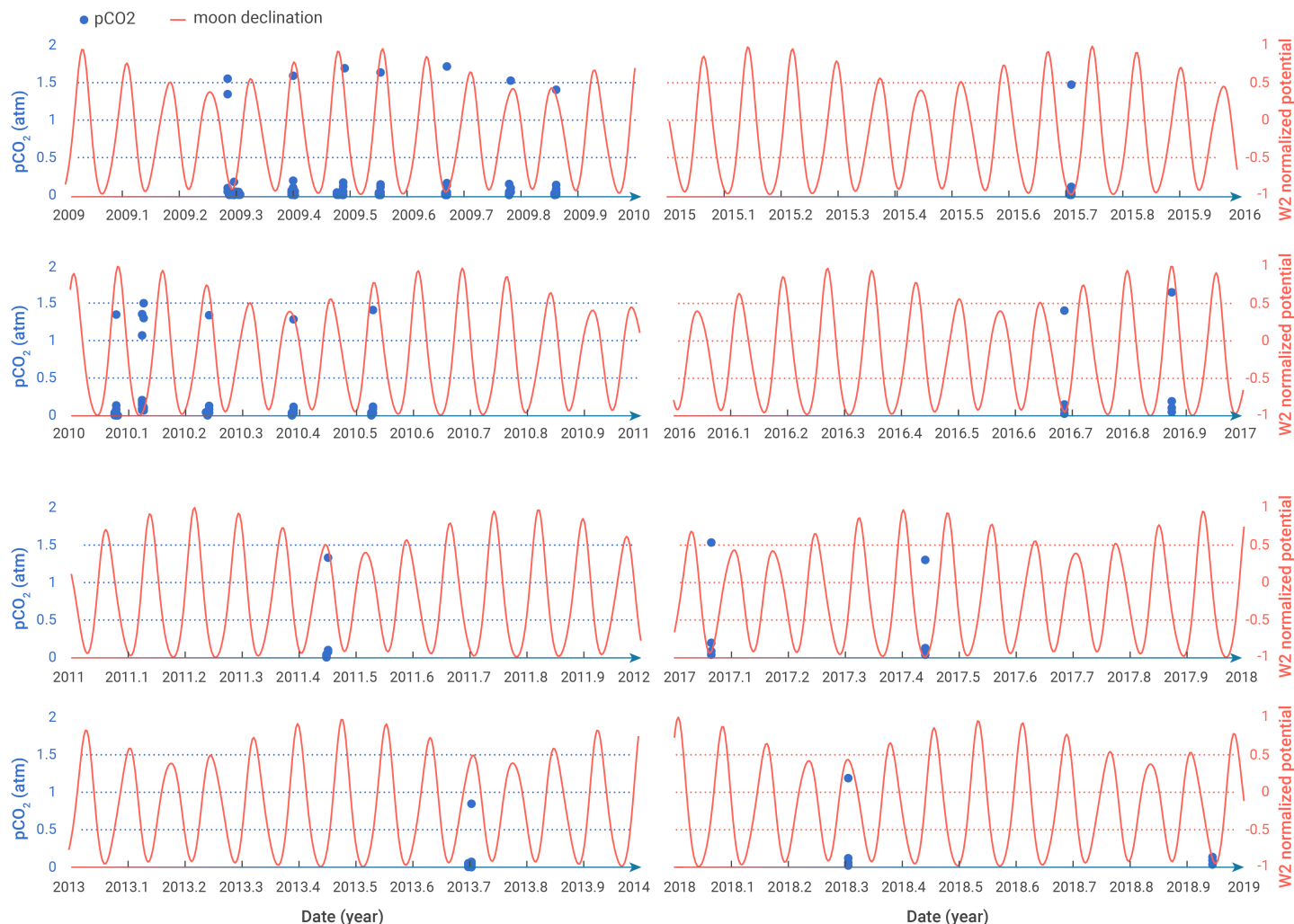
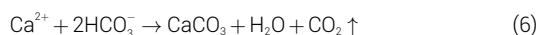


Figure 2. Evolution of pCO₂ partial pressure and Lunar tide potential estimated by Eqn.5 during the 10 years of investigation.

increased calcite precipitation maintaining calcite in near-equilibrium conditions. Additional processes such as halite dissolution from evaporate formation may effect groundwater composition in the studied zone, given that clay-water equilibria may contribute to the observed continuous pH variations.

However, tidal forces are too weak to fracture rock compared to tectonic forces.³⁰ We have estimated the vertical force acting on a unit mass at the average latitude for the investigated zone and found that the average force is close to $5.6 \times 10^{-7} \text{ N Kg}^{-1}$. Given the pressure gradient associated with tidal potential, the pressure between the surface and the first 10-15 Km (i.e. the transition between ductile and brittle crust) is approximately 10 Pa for an average rock density of 2.4 Kg m^3 . This value is lower by several orders of magnitude than the 0.1-0.20 MPa required to fracture carbonate rock in tensile stress.³¹ 10 Pa of tidal pressure may however drive fluid flow in stressed localized areas, increasing the movement of subsurface fluids.⁴ Groundwater at different depths may therefore be affected by tidal forces³² and carry with it the gas accumulated in buried highly pressurized aquifers as it travels towards the surface. At the fault planes, fluid pressure is reduced and local aquifers may connect favoring CO₂ release³³ with consequent carbonate precipitation, in agreement with the following the classical reaction path:



The formation of calcium carbonate deposits evidenced by the large quantities of travertine in the zone and the significant release of CO₂ point to disequilibrium conditions generated by a reduction in CO₂ pressure.^{18,34} When tidal potential is high and the Moon is close, aquifer depths are highest and fluids

are oversaturated with respect to calcite; whereas when tide potential is low, fluids are close equilibrium with calcite. Although lunar tides exert only weak stress on rock³⁵ without fracturing it,³¹ they do indeed produce weak vertical fluid movements^{18,34} causing ex-solution from CO₂-rich waters. This process, similar to the mechanical bubbling that occurs when you shake a bottle of carbonated soda, modifies the system's overall chemical equilibrium³⁶ and generates fluid releasing processes. The observed variations in central Apennine CO₂ release may correspond to gas separation and the ascent of CO₂ through fluid movements triggered by the lunar declination.

As confirmed by our investigation, tides are harmonic functions characterized by amplitudes and phases that simultaneously act as stressors on the subsurface, the contribution of lunar tides on the groundwater response can now be disentangle. We propose that systematically exploring the influence of subsurface properties on the tidal transfer function, the relationship between the amplitude and the phase of the stressors and the resulting chemical transport reactivity responsible for CO₂ releasing could be potentially predicted.

CONCLUSIONS

Our observational scale work shows the relation between lunar tides and CO₂ releasing in over-pressured CO₂ aquifers. The pressurized carbonate fluids rise to the surface through pre-existing faults producing variable amounts of CO₂ that are potentially released into the atmosphere. We propose that carbonate mineral dissolution under calcite near-equilibrium conditions may be responsible for the observed CO₂ variations in the Central Apennines and that changes in fluid dynamics can be triggered by periodical tidal potential variations.

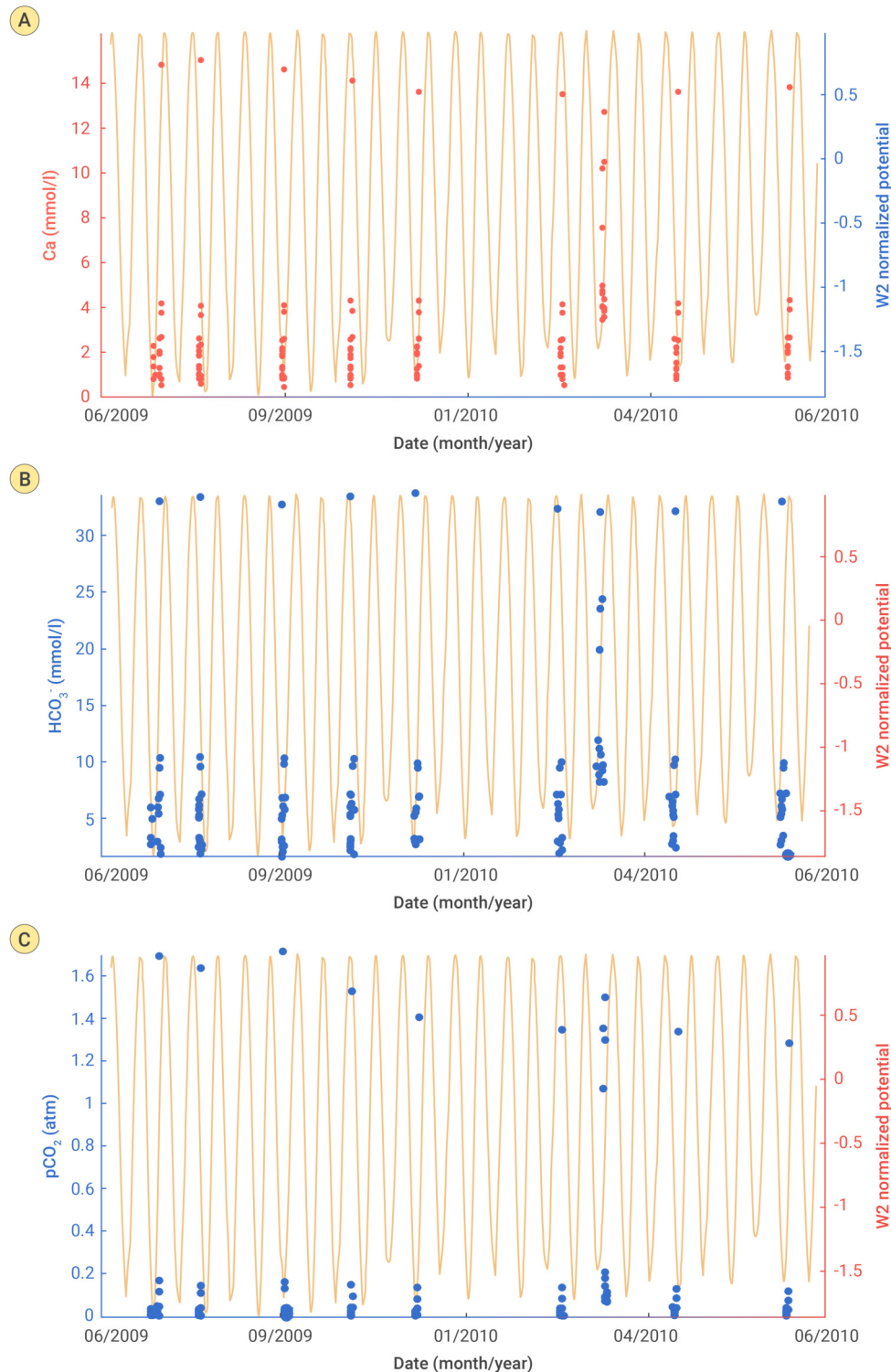


Figure 3. Evolution of calcium, HCO₃⁻, and pCO₂ over time (A) Evolution of calcium concentration (red dots) as a function of time for the year: June 2009–June 2010. The continuous black line corresponds to the continuous W potentials estimated by Eqn. 5. (B) Evolution of HCO₃⁻ (alkalinity) (blue dots) as a function of time for the year: June 2009–June 2010. The continuous black line corresponds to the continuous W potentials estimated by Eqn. 5. (C) Evolution of pCO₂ partial pressure (blue dots) as a function of time for the year: June 2009–June 2010. The continuous black line corresponds to the continuous W potentials estimated by Eqn. 5.

The analysis of long-term time series of CO₂ discharge and chemical fluid composition variations show that periodical low values of CO₂ released are correlated to the minimum lunar periodical tides of the zone. The combination of tidal potentials with traditional reactive transport description constraining poro-elastic and hydraulic parameter space can be potentially integrated in future predictive modelling of the CO₂ releasing from natural and artificial over pressured CO₂ reservoirs.

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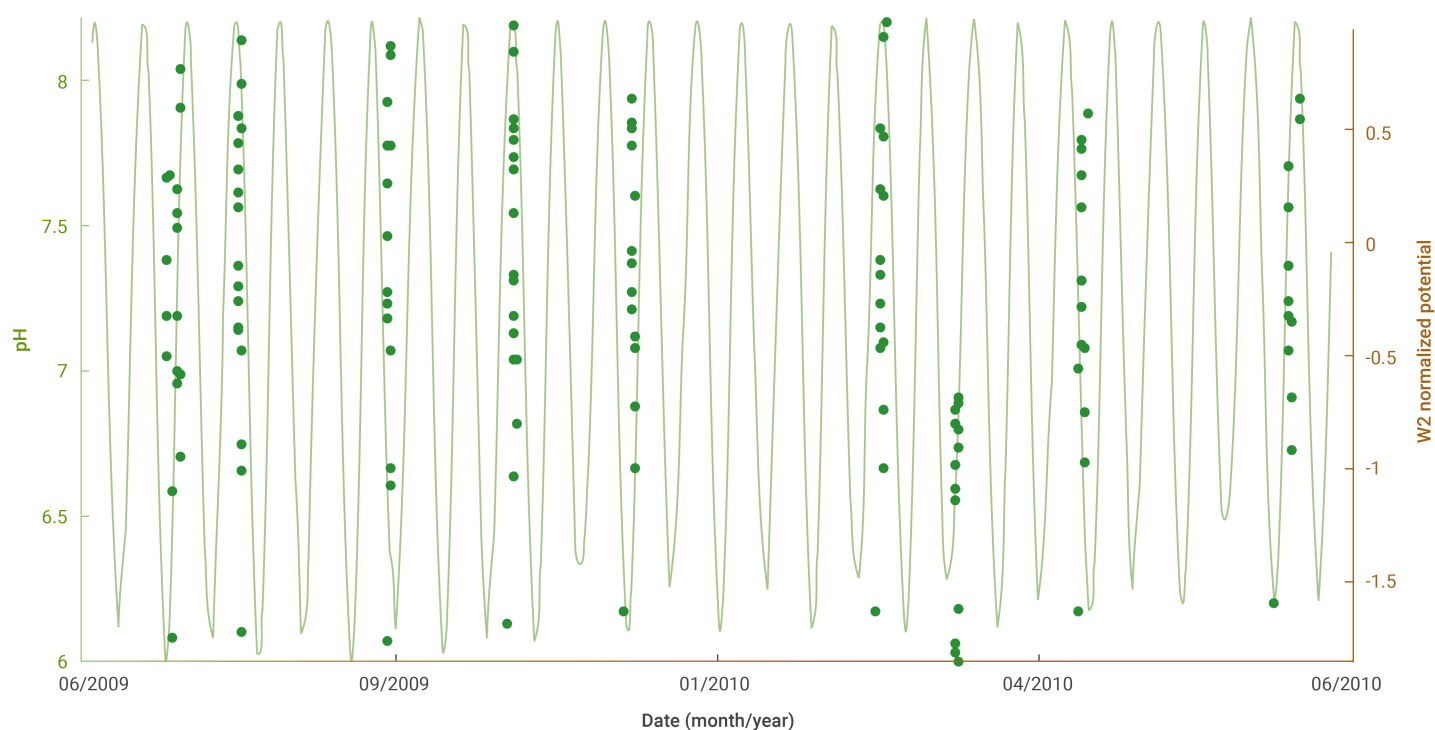


Figure 4. Evolution of pH (green dots) as function of time for the year: June 2009–June 2010 The continuous black line corresponds to the continuous W potentials estimated by Eqn.5.

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AUTHOR CONTRIBUTIONS

P. Z. and F. L. conceived and performed this manuscript.

DECLARATION OF INTERESTS

The authors declare no competing interests.

DATA AND CODE AVAILABILITY

All data and code are available from the corresponding author upon reasonable request.