Correlation between tectonic CO$_2$ Earth degassing and seismicity is revealed by a 10-year record in the Apennines, Italy

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Deep CO$_2$ emissions characterize many nonvolcanic, seismically active regions worldwide, and the involvement of deep CO$_2$ in the earthquake cycle is now generally recognized. However, no long-time records of such emissions have been published, and the temporal relations between earthquake occurrence and tectonic CO$_2$ release remain enigmatic. Here, we report a 10-year record (2009–2018) of tectonic CO$_2$ flux in the Apennines (Italy) during intense seismicity. The gas emission correlates with the evolution of the seismic sequences: Peaks in the deep CO$_2$ flux are observed in periods of high seismicity and decays as the energy and number of earthquakes decrease. We propose that the evolution of seismicity is modulated by the ascent of CO$_2$ accumulated in crustal reservoirs and originating from the melting of subducted carbonates. This large-scale, continuous process of CO$_2$ production favors the formation of overpressurized CO$_2$-rich reservoirs potentially able to trigger earthquakes at crustal depth.

INTRODUCTION
The relation between seismicity and fluid circulation has been demonstrated considering both the potential role of deep fluids in earthquake generation and the close spatial correlation between seismic zones and areas of deep fluid discharge (1, 2). The role of deep fluids in earthquake triggering has been recognized in different tectonic settings, including compressional (3–5), strike-slip (6, 7), and extensional (8–12) regimes. Besides, the involvement of CO$_2$-rich fluids in seismic sequences has been proven (6, 9–14). A worldwide spatial correspondence between seismically active areas and CO$_2$ emissions has been demonstrated (15, 16) as well as the primary role of extensional tectonics on Earth degassing of CO$_2$ (2). The few measured cases, Central-Southern Italy (17, 18) and Eastern African rift (19, 20), indicate the global relevance of tectonic CO$_2$ fluxes, which are thought to have controlled the CO$_2$ concentration of the atmosphere and thus of the climate (21).

The temporal variation of the rate of tectonic CO$_2$ discharge with seismicity is, however, still unconstrained because long time series of CO$_2$ discharge in seismically active areas have not been collected. Here, we show a 10-year-long (2009–2018) record of tectonic CO$_2$ emissions compared with seismicity in Central Apennine (Italy), an area where devastating historical earthquakes have occurred [e.g., the 1461 $M_w$ (equivalent magnitude) 6.4 and 1703 $M_w$ 6.7 events (22); http://storing.ingv.it/cfti/cfti5/]. The 2009–2018 observation period is a period of repeated seismic sequences (Fig. 1) that are considered here as a unique 10-year-long sequence (Central Apennine Seismic Sequence, hereinafter referred to as CASS). CASS includes the devastating main shocks of 6 April 2009 [$M_w$ (moment magnitude) = 6.3, L’Aquila earthquake], 24 August 2016 ($M_w$ = 6.0, Amatrice earthquake), and 30 October 2016 ($M_w$ = 6.5, Norcia earthquake). These earthquakes and their stronger aftershocks destroyed over a large area of Central Italy with more than 600 deaths, 2000 injured, and evacuation of about 120,000 persons. The three major earthquakes occurred at similar depths (8 to 12 km) with thousands of aftershocks concentrated in the upper 10 to 12 km of the crust. These shocks were characterized by normal focal mechanisms with a northwest-southeast (NW-SE) rupture strike consistent with that of the major Apennine faults (11, 23) that move in response to a northeast-southwest (NE-SW) extension. In the Apennine, crustal extension and devastating earthquakes occur in the overriding plate in response to regional uplift related to an upwelling and eastward-moving mantle wedge (17, 24–27). Our 2009–2018 degassing record of tectonic CO$_2$ flux shows that more than 1800 kt of deeply derived CO$_2$ were released from a relatively small area (~700 km$^2$) with a rate varying in time accordingly to the evolution of the seismicity.

RESULTS
Record of tectonic CO$_2$ emission compared with seismicity
Abundant, diffuse CO$_2$ degassing affects the western side of the Apennine chain (17) and confirms the occurrence of melting of subducting plate carbonates in the mantle (25). The upward migration of CO$_2$-rich fluids generates two large degassing structures at the surface: the Tuscan Roman degassing structure (TRDS) and Campanian degassing structure (CDS) that extend from the Tyrrhenian Sea to the Apennines (Fig. 1) (17). The seismic activity (http://terremoti.ingv.it/) mainly concentrates in the axial sector of the chain, at the eastern boundary of TRDS and CDS (Fig. 1). This intriguing spatial distribution, in light of the degassing structures, suggests that the gas, accumulated in buried highly pressurized structural traps, may favor seismicity (17, 27), because high-pressure CO$_2$ at depths can promote fault weakening mechanism (9, 28). CASS began at the eastern border of the TRDS (Fig. 1), where a deep source supplying endogenous carbon dioxide to the groundwater is detected.

The geochemical investigations aimed to detect and quantify the possible earthquake-related emission of deep CO$_2$ started immediately after the April 2009 mainshock (29); in particular, we focused...
on the CO$_2$ dissolved in two large carbonate aquifers close to the epicentral areas (Gran Sasso and Velino aquifers; Fig. 1B). To date, the geochemical dataset includes 270 chemical and isotopic compositions of spring waters sampled from April 2009 to December 2018 (see Materials and Methods). These samples were collected at the main 36 springs (fig. S1) whose flow rate amounts to ~70% of the total discharge of the two aquifers (~80 m$^3$ s$^{-1}$).

Concentration of deeply derived CO$_2$ ($C_{\text{deep}}$) is computed from the total dissolved inorganic carbon (TDIC) content of the water samples by applying a carbon mass balance (30) based on the carbon isotopes (see Materials and Methods). Remarkable variations are observed for three springs of Velino aquifer that were monitored for the entire period (Peschiera, Canetra, and San Vittorino), while the five springs of the Gran Sasso aquifer, monitored until 2015, show either a weak signal or no substantial variation in $C_{\text{deep}}$ when compared to the Velino springs (fig. S2). $C_{\text{deep}}$ of these latter springs follows the temporal evolution of the seismicity (Fig. 2, A to C). Peak values of $C_{\text{deep}}$ occur concurrently with the main shocks [i.e., the devastating events of $M_w$ 6.3 April 2009 and $M_w$ 6 and $M_w$ 6.5 August to October 2016; Fig. 2], and then CO$_2$ emissions decrease following the seismicity decay in terms of magnitude and rate. The robustness of the correlation between CO$_2$ and seismicity was investigated by testing the null hypothesis of no correlation (Pearson’s correlation coefficient equal to 0) through the Student’s $t$ distribution (31). The $C_{\text{deep}}$ is compared with the earthquake number, and the seismic energy release occurred at variable distance from the springs in a period centered at the sampling date ± a time lag. The correlation is statistically significant (significance level, 0.01) for a wide range of values for time lag (>10 days) and distance (20 to 80 km) (see Materials and Methods and fig. S3). This correlation is also well highlighted in the binary plots of Fig. 2 (A to C), where the $C_{\text{deep}}$ content of the three springs is compared with the number of earthquakes for a time lag of 40 days, at a distance <45 km. It is worth noting the high correlations of the earthquake number with the $C_{\text{deep}}$ of San Vittorino spring (the water that showed the strongest variation, $R^2 = 0.80$) and Peschiera spring (the spring of highest flow rate, $R^2 = 0.62$).

Although geochemical anomalies in groundwaters associated with CASS were already reported in many studies [e.g., (32–34) and references therein], we produce an unprecedented record of tectonic CO$_2$ emission (Fig. 2E) by applying a carbon mass balance to the Velino aquifer springs of known flow rate (see Materials and Methods). The daily flux of deeply derived CO$_2$ ($F_{\text{CO2}}$), similarly to $C_{\text{deep}}$ of the springs, shows a statistically significant correlation with seismicity (significance level, 0.01; see Materials and Methods and fig. S3). The total cumulative amount of the CO$_2$ released during the CASS period (10 years) is of ~1800 kt, i.e., of the same order of magnitude of the CO$_2$ involved in volcanic eruptions [see Table 2 in (35)]. It is worth noting that this amount is the minimum estimate of the total CO$_2$ involved, because it does not include neither the degassed fraction of CO$_2$ (i.e., part of the CO$_2$ is directly emitted in the atmosphere by gas emissions; see movie S1) nor the gas emitted in other areas.

**Origin of the CO$_2$**

Decarbonation triggered by frictional heating in the slip zone of carbonate-hosted faults has been suggested as a possible CO$_2$ release mechanism during earthquakes [e.g., (36, 37)]. However, this is not the case for CO$_2$ at the Velino aquifer, because the thermometamorphic CO$_2$ would have a very positive carbon isotope signature, as computable by theoretical fractionation and measured in laboratory experiments [$\delta^{13}\text{C} > +3$ per mil (%o)]. Instead, the Velino waters show the input of CO$_2$ with a slightly negative carbon isotope signature ($\delta^{13}\text{C} \sim −1.5$%o) during the entire observation period (Fig. 3), therefore excluding a major contribution of CO$_2$ formed by frictional heating during the earthquakes.

The gas source must be sought in the westward subduction of carbonates of the Adriatic plate beneath the Tyrrenhian mantle. According to (25), melting of such carbonates, due to the high temperatures at the interface between the mantle wedge and the subducting plate, generates carbonate-rich melts. These low density and viscosity melts can migrate upward through the mantle causing zones of partial melting (and CO$_2$-rich compositions) whose presence at depths from 130 to 60 km is revealed by shear-wave velocity images (25). The lower pressures at shallower depths (<60 km) may induce massive CO$_2$ degassing and accumulation beneath the Moho.
The presence of a crustal CO₂ reservoir at 10- to 15-km depth in the L’Aquila basin, where the CASS started in 2009, is indicated by a sill-like negative anomaly of P-wave seismic velocity (V_p) (Fig. 4) and low ratio of P and S-wave seismic velocities (V_p/V_s) zone [see Fig. 4 in (38)]. This zone was interpreted as a CO₂ storage zone (39) fed by fluids from the underlying mantle wedge. The feeding process explains the high geothermal advective heat fluxes (~200 to 300 mW m⁻²) transmitted through the aquifers located in the area (39). The deep CO₂ storage zone is underneath a great portion of the Velino aquifer (Fig. 4), where CO₂ emision has been documented. It is worth noting that the total thermal energy associated with the CO₂ emission during 2009–2018 is two orders of magnitude larger than the total seismic energy released by CASS (~6 x 10²³ versus ~6 x 10²¹ erg; see Materials and Methods).

The deep origin of the tectonic CO₂ detected at Velino aquifer is also in agreement with previous seismological studies that highlighted the primary role of deep fluids in triggering and controlling the temporal evolution of CASS and of previous seismic sequences (10, 11, 40, 41). For example, gas pulses from deep, highly pressurized reservoirs are recognized as the main source of the 1997 Umbria-Marche aftershocks (9) and of the 2013 Sannio-Matese earthquakes, these latter related to gas release from a magma intrusion in the lower crust (12).

DISCUSSION
In the Apennine chain, the crustal CO₂ reservoirs such as that located below the Velino aquifer (Fig. 4) are fed by the ascent of CO₂-enriched slab-derived fluids (25), a process implying a continuous mechanism of CO₂ production and accumulation in the crust. This ceaseless movement of CO₂ from depth causes pressurization in the reservoir and consequent transfer of gas to the uppermost crustal layers and, ultimately, CO₂ saturation-oversaturation of the overlying aquifer(s). At regional scale, this mechanism explains the formation of the large CO₂ degassing structures (TRDS and CDS in Fig. 1) and the formation of numerous emissions of free gas (Fig. 1).
The occurrence of CO₂ saturated-oversaturated waters at the Velino basin is testified by the Terme di Cotilia springs, which discharge hundreds of liters per seconds of CO₂ oversaturated waters, and by the direct emissions of a CO₂-rich gas phase (movie S1). Different mechanisms have been proposed for the transfer of CO₂-rich fluids from depth to surface. These include the dynamic, long-term fluid pumping of fluids related to grain-size reduction through the nucleation phase in the ductile mantle-crust shear zone (42), the ascent of fluids along deep-seated faults (43), the fluid pressurization by self-sealing processes and release (fault-valve) during earthquakes at the base of the seismogenic layer of the crust (44), and the emplacement of magma within the crust (12). Here, we propose that, in addition to the above processes, seismic shaking causes sudden ascent of already separated gas bubbles and, possibly, new gas exsolation from the CO₂-rich solutions (mechanical bubbling such as a shaken bottle of a carbonated drink). This process of fluid release is consistent with (i) the observed, almost simultaneous, increase of deep CO₂ with earthquakes at the surface (Fig. 2) and (ii) the occurrence at depth of earthquakes triggered by a fluid-related pressure front as suggested by the diffusive spatiotemporal evolution of the earthquakes (11). The increase of tectonic CO₂ flux at the surface can be caused by the gas released at shallow depths, while the formation of fluid-related pressure fronts would be explained by the gas separated at depth and by the ascent of increasing amounts of CO₂ from the deep reservoir. It is worth noting that this process implies a feedback because seismicity causes gas separation with the formation of ascending pressurized gas fronts that, in turn, favor seismicity.

The ascent of fluids in tectonically active zones may be a passive process favored by fracturing of the crust during earthquakes and/or an active process in which pressurized fluids rise to the surface along preexisting faults, thus triggering earthquakes. Taking into account (i) the close relationship between CO₂ discharge and rate/magnitude of the 2009–2019 CASS earthquakes (Fig. 2), (ii) the diffusive processes of seismicity migration and rate (11), (iii) \( V_P \) and \( V_P/V_s \) tomographic models (45), (iv) variations in fluid pore pressure during the preparatory phase of the 2009 and 2016 mainshocks (46), and (v) the fluid pressure larger than the regional minimum horizontal stress in the upper crust during the Apennine seismic sequences (10, 11), we propose that the influx of CO₂ modulates the evolution of CASS.

We conclude that the ascent of huge amount of slab-derived CO₂ that continually accumulates at depth may significantly contribute to earthquake occurrence in the Apennines and, potentially, to the observed uplift (47) of the chain. The analysis of long-term time series of CO₂ discharge as presented here should be extended to other seismically active areas to better constrain the role of fluids
in modulating the seismic activity and to estimate the overall tec- 
tonic CO₂ flux on Earth.

MATERIALS AND METHODS

Field and laboratory measurements of groundwater

The complete dataset of the waters used in this work is reported in 
data file S1, while their location is reported in fig. S1. The first 
survey was performed in 1997, while the other surveys were carried 
out after the 2009 L’Aquila earthquake. Overall, 36 springs were 
sampled. Among these, 14 springs were monitored from 2009 to 
2015: 4 springs belong to the Velino carbonate aquifer (total dis- 
charge Q tot = 30.0 m³ s⁻¹; sampled discharge Q s = 24.75 m³ s⁻¹), and 
10 springs belong to the Gran Sasso carbonate aquifer (Q tot = 49 m³ s⁻¹; 
Q s = 25.6 m³ s⁻¹). The Velino aquifer springs, which showed re- 
markable variations, were sampled other six times after the earth- 
quake of August 2016. Data collected in 1997 and from April 2009 
to February 2010 were previously published (29, 30) (see data file S1).

For each spring, temperature, pH, Eh, electrical conductivity, 
and total alkalinity were determined directly in the field. The alka- 
linity determination was performed by acid titration with 0.01 N 
HCl. Water samples for chemical analyses were filtered with 0.45-μm 
filters and collected in three 100-ml polyethylene bottles. One aliquot 
was immediately acidified with HCl 1:1 diluted.

For the determination of δ¹³C of TDIC (δ¹³C TDIC ), the dissolved 
carbon species were precipitated in the field as SrCO₃ by adding 
SrCl₂ and NaOH in solid state to the water sample. Carbonate pre- 
cipitates were then filtered and washed with distilled water in a 
CO₂-free atmosphere in the laboratory.

Dissolved ions were analyzed at the laboratory of Earth Science 
Department of Perugia University. Dissolved anions (Cl, F, SO₄, 
and NO₃) were determined by ion chromatography using a Dionex 
DX-120. Ca and Mg were determined by atomic absorption flame 
spectroscopy on the acidified sample, while Na and K were deter- 
mined by atomic emission flame spectroscopy. All the laboratory 
analytical methods have an accuracy better than 2%.

Analyses of water hydrogen and oxygen stable isotopes (δD and 
δ¹⁸O) and of δ¹³C TDIC were performed at INGV (Istituto Nazionale 
di Geofisica e Vulcanologia, Sezione di Napoli), using a Finnigan Delta 
plus XP continuous flow mass spectrometer coupled with a GasBench 
II device. Each sample was analyzed in replicate. Carbon samples were 
analyzed versus the Working Standard Marmo Acqua Bianca (MAB) 
(carbonate rocks from the investigated aquifers [+1.8‰; (29, 30)), the 
carbon concentration of the infiltrating water), whereas C ext also 
originates from the addition of deeply derived CO₂ (C deep ) to normal 
groundwater in groups B and C (Fig. 5). The group C waters can be 
differentiated from group B given its higher amounts of added C deep 
that causes CO₂ degassing (P CO₂ ~1 bar in group C waters), calcite 
precipitation, and isotopic fractionation during the carbon removal.

Carbon mass balance

To quantify the contribution of the different carbon sources and to 
characterize their isotopic composition, we compute for each sample 
C ext and δ¹³C ext using the following carbon mass balance equations

\[ C_{\text{ext}} + C_{\text{carb}} = \text{TDIC} \]  
(1)

\[ \delta^{13}C_{\text{ext}} \times C_{\text{ext}} + \delta^{13}C_{\text{carb}} \times C_{\text{carb}} = \delta^{13}C_{\text{TDIC}} \times \text{TDIC} \]  
(2)

where (i) TDIC and δ¹³C TDIC are analytically determined; (ii) C car - 
b is computed as the sum of Ca + Mg + SO₄ considering the dissolu-
tion of carbonate minerals (i.e., calcite and dolomite) and the possible 
presence of gypsum/anhydrite; and (iii) δ¹³C car is assumed to be 
constant and equal to the average δ¹³C of numerous samples of 
carbonate rocks from the investigated aquifers [+1.8‰; (29) and 
references therein].

The two equations can be used to determine C ext and δ¹³C ext only 
for waters that have not experienced CO₂ degassing and calcite 
precipitation, a condition that, for the studied aquifers, is verified for 
TDIC <40 mM (29), i.e., only for the samples of group A and 
group B (Fig. 5A).

The concentrations (C inf and C deep) and isotopic compositions 
(δ¹³C inf and δ¹³C deep) of the carbon from the different sources 
contributing to C ext of group B samples are computed by considering 
the following carbon balance equations

\[ C_{\text{inf}} + C_{\text{deep}} = C_{\text{ext}} \]  
(3)

\[ \delta^{13}C_{\text{inf}} \times C_{\text{inf}} + \delta^{13}C_{\text{deep}} \times C_{\text{deep}} = \delta^{13}C_{\text{ext}} \times C_{\text{ext}} \]  
(4)

The system of two equations and four unknown variables is 
solved for each sample using the binary plot δ¹³C ext Versus 1/C ext 
(Fig. 5B) where mixtures among different sources show a linear 
trend. In particular, in the case of the springs of Velino aquifer:

(i) The isotopic compositions of the deeply derived CO₂ 
(δ¹³C deep = −1.48‰) is considered unique and derived in Fig. 5B as 
the δ¹³C ext intercept at 1/C ext = 0 (i.e., pure CO₂ end member) of 
the best-fit linear regression (δ¹³C ext = −1.48 − 28.1/C ext) of the samples; 
(ii) C inf is determined at the interception of the line connecting 
each sample to the deep source (1/C ext = 0, δ¹³C ext = −1.48‰) with 
the infiltrating water line (δ¹³C inf = −25.0 + 13.2/C inf) computed as 
the best-fit linear regression of group A (normal groundwater). This 
sample by sample computations gives a narrow range of C inf val-
ues, from 1.5 to 2.18 mM with a mean C inf = 1.76 ± 0.18 mM;
(iii) the carbon concentration of the deeply derived CO₂ (C_{deep}) of each sample (Fig. 2, A to C) is given by inserting the computed mean C_{inf} in Eq. 3.

Note that, to have a minimum estimate of the C_{deep}, a simplified computation is applied also to the group C Terme di Cotilia spring, where part of the carbon is removed during degassing and calcite precipitation. In details, we compute C_{ext} from Eq. 1 and C_{deep} from Eq. 3, assuming the mean C_{inf} value derived for the group B waters.

**Total CO₂ and energy output from Velino aquifer**

The total deeply derived CO₂ entering the Velino (F_{CO2}) is computed by summing the contributions of the monitored springs and applying a correction to account for the portion of the nonsampled groundwaters. The different contributions are computed by multiplying the C_{deep} of the four monitored springs for a corresponding water discharge. For this computation, we consider the Peschiera, Canetra, Terme di Cotilia, and San Vittorino springs as representative of the discharge from groups of springs of similar compositions sampled in the detailed survey of January–February 2010. We consider a flow rate of 18 m³ s⁻¹ for Peschiera, 6 m³ s⁻¹ for the Canetra group, 0.25 m³ s⁻¹ for Terme di Cotilia, and 0.5 m³ s⁻¹ for the San Vittorino group according to data from detailed hydrogeological studies (49, 50). Last, we scale the computed total CO₂ of the monitored springs (24.75 m³ s⁻¹) to the total discharge of the aquifer (i.e., 30 m³ s⁻¹).

The total energy associated with the deep CO₂ influx in the Velino aquifer is estimated starting from the good correlation between the deep CO₂ and the advective geothermal heat computed by (39) and (18) for 11 aquifers of the Central Apennine [which includes also the Velino aquifer, named Marscia N in (39)]. The good correlation suggests that the gas and the heat are advectively transported into the aquifers by hot CO₂-rich fluids. The estimated CO₂ flux from the data of each survey was used to estimate the thermal energy by applying the relation between the F_{CO2} (in t day⁻¹) and the geothermal heat flow (Q_H in erg day⁻¹), Q_H = 3.3 × 10¹⁷ F_{CO2}, computed by the data reported in (18).

**Correlation between deep CO₂ and seismicity**

To track the deep CO₂, we consider four time series: C_{deep} from Peschiera, Canetra, and San Vittorino springs and F_{CO2}. The earthquake catalog includes seismic events with M_w >2.0 from September 2007 to August 2019. We verified the completeness of the catalog for this period and magnitude range. For each C_{deep} value, we extract from the catalog earthquakes within a given time lag before and after the sampling date and within a given distance from the corresponding spring (from Peschiera spring in the case of F_{CO2}), evaluating the number and the energy release (51) of the extracted events. In this way, for each C_{deep} and F_{CO2} time series and each couple time lag–distance, we build two seismic time series considering the logarithm of the earthquake number and of the total energy release.

We test the correlation between each C_{deep}–F_{CO2} series and each corresponding seismic time series (log number and energy) for a wide range of time lags (from 7 to 70 days) and distances (from 15 to 100 km). In each case, we test the null hypothesis of no correlation (Pearson’s correlation coefficient equal to 0) through the Student’s t distribution (31). As shown in fig. S3, the null hypothesis of no correlation can be rejected at significance levels of 0.05 and 0.01 for all time lags (>15 days) and distances (>30 km), with the exception of the case of C_{deep} at Canetra. In this specific case, the null hypoth-

**SUPPLEMENTARY MATERIALS**

Supplementary material for this article is available at http://advances.sciencemag.org/cgi/content/full/6/38/eabc2938/DC1

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