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Key Points:

- We present the world's first time series acquired with an absolute quantum gravimeter in the summit crater zone of an active volcano
- Despite the unfavorable ambient conditions, the quantum gravimeter produced high quality data, allowing to track volcano-related changes
- Comparison with the data from other gravimeters highlights changes due to bulk mass redistributions within Mt. Etna's feeding system

Supporting Information:

Supporting Information may be found in the online version of this article.

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Detecting Volcano-Related Underground Mass Changes With a Quantum Gravimeter

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Abstract We present the world's first time series acquired in the summit area of an active volcano with an absolute atom interferometry gravimeter. The device was installed ~2.5 km from the active craters of Mt. Etna volcano and produced a continuous high-quality gravity time series, despite the unfavorable environmental conditions at the installation site and the occurrence of phases of high volcanic tremor during the acquisition interval. Comparison with data from superconducting gravimeters installed elsewhere on Mt. Etna highlights correlated anomalies, demonstrating that the quantum device measured gravity variations driven by bulk mass changes. The latter are reflective of volcanic processes, involving the dynamics of magma and exsolved gas in the upper part of Mt. Etna's plumbing system. Our results confirm the operational possibilities of quantum gravimetry and open new horizons for the application of the gravity method in geophysics.

Plain Language Summary Mass redistributions occurring in the Earth's interior, for example, when a magma batch is displaced through the feeding system of an active volcano, may induce tiny changes in gravity over time, measurable on the ground. Measurement of such changes requires high-precision devices, namely, the gravimeters, which can detect variations as small as one part in 10⁹ of the gravity acceleration on Earth. However, standard gravimeters are not ideally suited for use in harsh field conditions, especially when continuous measurements are the target. Recent advances in quantum technology have allowed the development of a portable gravimeter which can successfully operate under field conditions. Here we present the world's first application of this quantum gravimeter for monitoring and studying an active volcano. The device was deployed only 2.5 km away from the summit active craters of Mt. Etna volcano (Italy) and has provided a high-quality gravity time series. Inspection of this time series highlighted gravity changes which are reflective of bulk volcanic processes, involving magma and exsolved gas in the upper part of Mt. Etna's plumbing system.

1. Introduction

In recent years, monitoring of subsurface masses, natural or engineered, has become increasingly important. The study of geological reservoirs can address issues related with energy supply, climate change mitigation and the assessment of natural hazards, such as seismicity and volcanic eruptions (Dzurisin, 2003; Poland & Anderson, 2020; Thierry et al., 2015). Understanding aquifer charge and discharge, carbon sequestration and volcanic processes involves multi-parametric approaches to inform predictive and simulation models. Gravimetry provides unique direct information on changes in the distribution of underground masses over time (Carbone et al., 2017; Van Camp et al., 2017). It can therefore be utilized to gain insight into the dynamics of subsurface fluids, like water, hydrocarbons, and magma (Chapman et al., 2008; Forster et al., 2021; Gasperikova & Hoversten, 2008; Sugihara & Ishido, 2008). However, gravimetry has not been employed extensively, mainly due to stringent instrumental limitations. The application of gravimetry to geophysical studies requires that gravity changes between one part in 10⁶ and one part in 10⁹ of the standard gravity on Earth ($g = 9.8 \text{ m/s}^2$) should be precisely measured over timescales between minutes and years and at several locations. This is only marginally compatible with currently available instruments (Van Camp et al., 2017), which are, in general, difficult to utilize in the field, especially under the harsh environmental conditions characterizing geodynamically active areas (e.g., the active craters of a volcano). Spring gravimeters are compact, easily transportable and take little power to work. They can thus be deployed close to active structures. Nevertheless, these instruments suffer from strong instrumental drift, implying that they cannot be used to continuously track gravity changes over intervals longer than a few days. Superconducting gravimeters (SGs) are negligibly affected by instrumental drift and can thus precisely measure gravity changes occurring over time scales of minutes to years. However, they are difficult

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to transport, setup and, more importantly, require ~ 1.4 kW of continuous power to work, making them mostly unsuited for installation under unfavorable field conditions. Quantum technologies provide a key to address this challenge. Sensors based on matter-wave interferometry with cold atoms are now reaching a high level of maturity (Geiger et al., 2020). Technological developments have been driven by possible applications in future space missions, which led to experiments with quantum sensors in challenging environments (Barrett et al., 2016; Becker et al., 2018; Geiger et al., 2011; M ntinga et al., 2013). In parallel, the performances of high precision absolute quantum gravimeters were demonstrated in the laboratory (Freier et al., 2016; Gillot et al., 2014; Hu et al., 2013; Wang et al., 2018) and outside laboratories, using transportable prototypes (Bidel et al., 2018, 2020; Cooke et al., 2021; M noret et al., 2018; Wu et al., 2019).

Here, we report on a leap forward in quantum technology, by demonstrating a 4-month time series with a resolution of one part in 10^9 of g , acquired in the summit area of Mt. Etna volcano, through the absolute quantum gravimeter named AQG-B. Data from the AQG-B are compared with data from two superconducting gravimeters (SGs), installed at different locations on Mt. Etna (Carbone et al., 2019). Correlated gravity changes amongst the three time series point to mass changes beneath the summit craters area as the common source.

2. Deployment of the AQG-B at Mt. Etna

The AQG-B, produced by iXblue (formerly Muquans), is packaged in three main modules (Figure 1a) with reduced size and weight (below 300 L and 40 kg each): (a) a power supply module, managing electrical power and thermal regulation, (b) the laser system, generating the laser light required to manipulate the atoms, and (c) the sensor head where the atomic measurements take place. Thanks to thermostatic controls in the laser system and sensor head, the device features an operating temperature range of 0–40°C. To ease field operation, the number of connections between the laser system and the sensor head has been reduced to only 4, with a cable (including the optical fiber) length of 15 m. The measurement sequence of the AQG-B is repeated at a frequency of 1.85 Hz (M noret et al., 2018; see Text S1 in Supporting Information S1).

Mt. Etna is one of the most active and better monitored volcanoes in the world (Cannav  et al., 2019). It has been the site of microgravity measurements with different types of instruments (Carbone et al., 2008, 2017, 2019). Only spring gravimeters could be deployed in the close vicinity of the active craters, where the strongest volcano-related gravity changes develop (Carbone et al., 2015). Between 2014 and 2016 a mini-array of iGrav SGs was installed at Mt. Etna (Carbone et al., 2019), at minimal distance from the summit active craters of about 3.5 km.

In the framework of the NEWTON-g project (Carbone et al., 2020), an AQG-B was deployed close to the summit of Mt. Etna, at the Pizzi Deneri volcanological observatory (PDN; 2,800 m elevation, 2.5 km from the summit craters; Figures 1a and 1c), for continuous acquisition. The PDN observatory is the closest building to the summit craters of Mt. Etna where long-term continuous gravity measurements can be performed. Nevertheless, harsh environmental conditions (strong daily and seasonal temperature variations, high levels of volcanic tremor, inaccessibility due to snowfall during the months of winter/spring) make the execution of high-precision gravity measurement especially challenging at this location.

The observatory includes two interconnected dome-shaped shelters on the ground floor and a large room in the semi-underground basement (Figures 1c and 1d). This room features a concrete pillar that was expressly designed for microgravity measurements. It is directly rooted in the rock underlying the building and detached from the building itself to avoid vibration transmission. Due to the reduced size of the stairway, only the sensor head of the AQG-B could be installed on the concrete pillar. We used the standard 15-m cable to connect it to the power supply and laser system, installed on the ground floor (Figure 1d). As mains electricity is not available and the site is often unreachable in winter due to snow cover, reliable power supply and data transmission systems are needed to allow for long-term unsupervised operations. In order to provide the continuous electrical power required by the AQG-B (within 500 W), a custom hybrid off-grid power system was installed at PDN (see Text S2 in Supporting Information S1). The existing Wi-Fi link of the observatory, managed by INGV-OE, allows for transmission of data and remote operation of the AQG-B through a dedicated software interface.

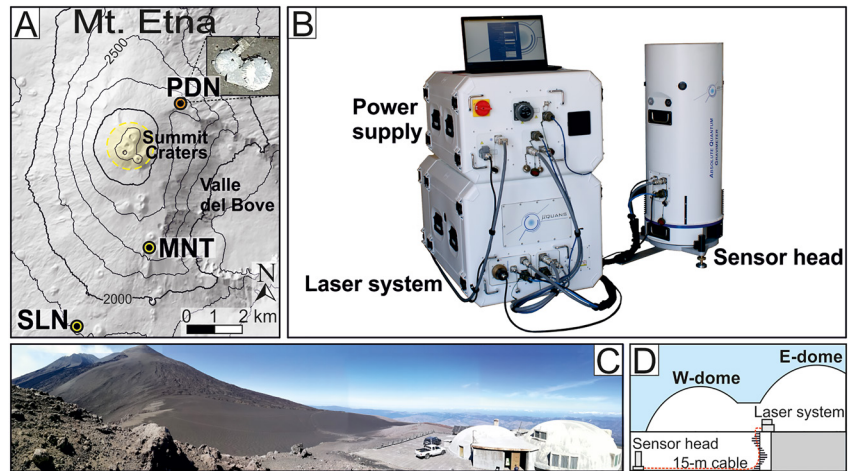


Figure 1. Deployment of the AQQ-B in the summit craters area of Mt. Etna volcano. (a) Sketch map of Mt. Etna showing the position of PDN, SLN, and MNT stations. The shaded yellow area encloses all the possible horizontal positions of the mass source (Figure 4 and text). Inset: aerial photo of the PDN observatory (Google Maps). (b) Picture of the AQQ-B. The height of the sensor head is 100 cm. (c) Panorama looking NW, from the slope to the S of the observatory, showing the proximity of PDN to the summit craters. (d) Section (not in scale) crossing the observatory and showing the installation configuration of the AQQ-B.

3. Rejection of Volcano-Related Ground Vibrations

The characteristics of seismicity at Mt. Etna, especially in the summit craters zone, are different from those at the quiet sites where gravity measurements are typically performed. In particular, much of the seismic energy is concentrated over the frequency band between 0.1 and 10 Hz (Saccorotti et al., 2004). This frequency range overlaps with the maximum sensitivity of the acceleration transfer function of the AQQ-B, which is a low-pass filter with cut-off at $1/2T \approx 8$ Hz (Geiger et al., 2011). We estimate that most of the noise on the signal from the AQQ-B is due to the vertical component of the ground motion, while the horizontal component has a negligible effect (Text S3 in Supporting Information S1). In order to mitigate the effect of vertical ground vibrations, we implement an active compensation system (Lautier et al., 2014). This was initially based on the use of a high-performance classical accelerometer (Nanometrics Titan) integrated in the sensor head (Ménoret et al., 2018). This sensor is particularly well adapted for high frequencies, but the high-pass filters required to remove the DC component of the sensor introduce a significant dephasing at low frequencies, where the amplitude of the signal is high at Mt. Etna.

Simulations based on (a) vibration spectra previously recorded at PDN under different levels of volcanic tremor and (b) the response functions of both the classical accelerometer and the atom interferometer, showed that, using the standard vibration compensation system, it is not possible to obtain a gravity signal-to-noise ratio compatible with the required performances.

The vibration rejection system was therefore optimized by replacing the accelerometer with a broadband seismometer (Nanometrics Trillium Compact 120s). New simulations showed that, with the improved vibration rejection system, the AQQ-B can reach sensitivities between 1,200 and 2,500 nm/s^2 at 1s, depending on the level of volcanic tremor.

Due to the lower cutoff frequency (around 100 Hz), vibration data acquired through the seismometer are more delayed (~ 2.5 ms) than data from the accelerometer (cutoff frequency = 430 Hz; delay of around 500 μs), implying that more data are missing when the phase correction is applied to the Raman laser. Combined with a higher sensitivity near the first and last pulses when using the velocity transfer function, this can significantly deteriorate the quality of the real-time compensation. We mitigate this effect by applying a post-correction to the atomic ratio, corresponding to the residual vibration phaseshift due to the uncompensated delay.

To quantify the efficiency of vibration rejection, we compare the distribution of interferometric phases computed from the seismometer signal to the distribution of phases at the output of the atom interferometer after real-time

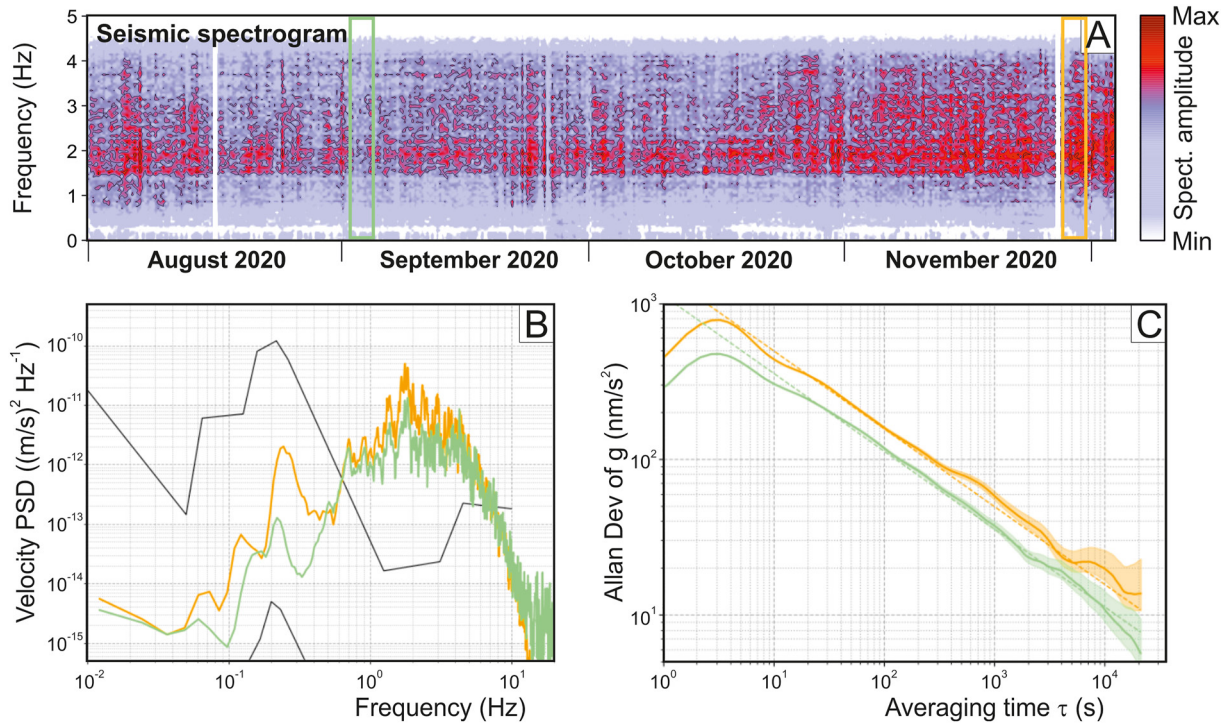


Figure 2. Sensitivity analysis on data from the AQG-B. (a) Spectrogram of the vertical-component seismic signal from the Pizzi Deneri station (EPDN seismic station; 1 August–3 December 2020). The green and orange boxes mark periods of low and high volcanic tremor amplitude, respectively. (b) Power spectral density of the seismic velocity recorded at EPDN, during the phases of low (green) and high (orange) tremor amplitude marked in the spectrogram. Black curves indicate the new high and low noise models from Peterson (1993). (c) Allan deviation of the corrected gravity data from the AQG-B at PDN, during the low (green) and high (orange) tremor phases. The green and orange dashed lines in the Allan plot indicate a sensitivity of 1,200 and 1,600 $\text{nm/s}^2/\tau^{1/2}$, respectively.

compensation. The resulting rejection efficiency is of the order of 100. In other words, if no active compensation was implemented, phase noise would have a standard deviation of several radians instead of a few tens of milliradians with the active compensation. This is beyond the linear range of the atom interferometer, meaning that, without the active compensation, it would not be possible to recover the value of gravity acceleration unambiguously (Lautier et al., 2014).

Figure 2a shows the spectrogram of the vertical component of the seismic signal from PDN during 1 August–3 December 2020. The green and orange boxes mark two subintervals of low and high tremor, respectively. Power Spectrum Densities of the seismic velocities during the two subintervals are shown in Figure 2b. Allan deviation analysis (Riley, 2008 and Text S4 in Supporting Information S1), performed on the corrected gravity data from the AQG-B at PDN (Figure 2c), indicates a sensitivity of approximately 1,200 and 1,600 nm/s^2 at 1 s, for the subintervals of low and high tremor, respectively, corresponding to only a 2 to 3-fold degradation with respect to a quiet laboratory environment (Carbone et al., 2020). These results show the efficiency of the real-time vibration rejection strategy relying on a broadband seismometer and suggest that, even during periods of high volcanic tremor, the data quality is suitable for studying volcano-related gravity changes. Indeed, once data from the AQG-B are averaged over time windows of 1–5 hr, it is possible to track gravity changes with amplitudes ranging between a few tens and a few hundreds of nm/s^2 , occurring over time scales of days to weeks (see Section 4 and Figure 3).

4. Results

Gravity data from the AQG-B are corrected for environmental effects (Earth tide, polar motion, atmospheric pressure, ground tilt) and possible instrumental drifts (see Text S5 and Figure S1 in Supporting Information S1). The instrument recorded gravity data at PDN for ~ 4 months (1 August–3 December 2020; Figure 3a). The gaps in the time series are due to temporary failures of the off-grid power supply system, which caused abrupt shutdowns of the AQG-B. These failures did not damage the instrument, and a remote restart of all the subsystems (ion pump,

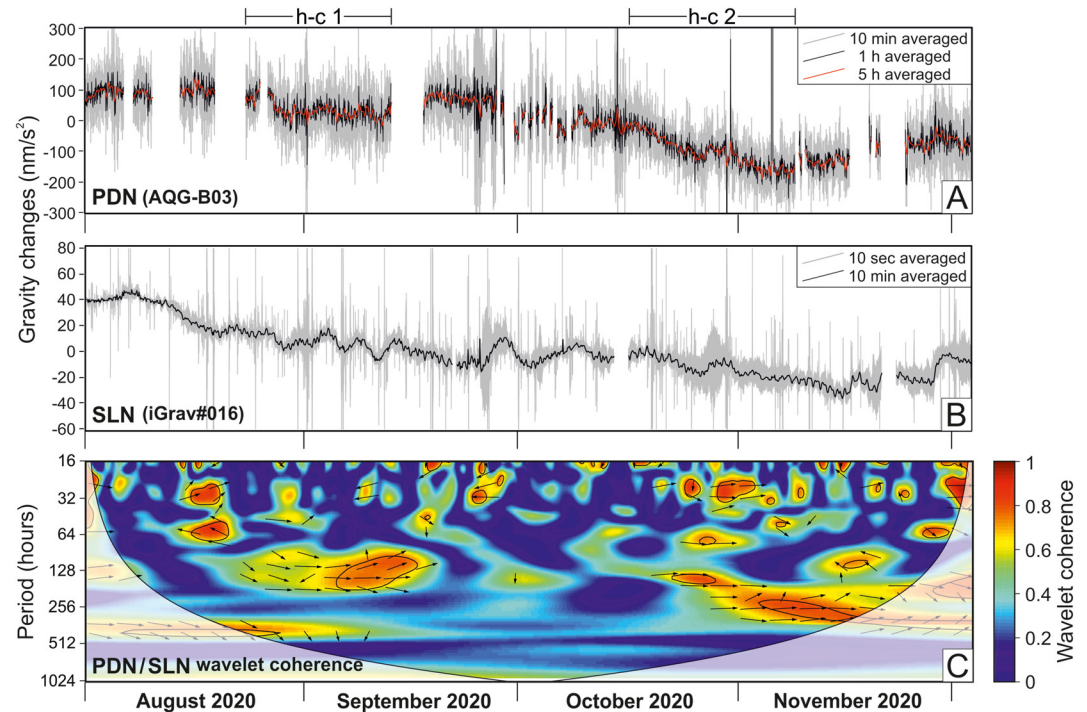


Figure 3. Gravity time series and wavelet coherence analysis. (a) Gravity time series from PDN (1 August–3 December 2020), after removal of the perturbing effects shown in panels (b–f) of Figure S1 in Supporting Information S1. Data are averaged over 10-min (gray curve), 1-hr (black curve) and 5-hr (red curve) intervals. (b) Gravity time series from SLN, after removal of Earth tide, atmospheric pressure, ground tilt and polar motion. Data are averaged over 10-s (gray curve) and 10-min (black curve) intervals. (c) Wavelet coherence between the signals in panels (a and b). Black arrows indicate the relative phase relationship (in-phase pointing right, anti-phase pointing left, and one signal leading the other by 90° pointing straight up or down). The cone of influence, where edge effects may distort the results, is shown as a lighter shade.

electronics, lasers) was possible once electrical power was restored. Thanks to the accuracy and repeatability of the AQG-B, no instrumental change in the average level of the signal occurred across any of the above gaps. Furthermore, no significant instrumental drift affects the data from the AQG-B during the studied period.

Residual gravity data from the AQG-B are cross-analyzed with data produced by the two iGrav SGs installed on the southern slopes of Mt. Etna (Carbone et al., 2019). Figure 3b shows the residual gravity signal from iGrav#16, installed in the facilities of the Serra La Nave astrophysical observatory (SLN; Figure 1a). The signal is corrected for the same external perturbing effects as the signal from the AQG-B. Thanks to the low-noise characteristics of SGs and also due to the fact that the instrument is installed relatively far from the active summit area of the volcano (~ 6.5 km), a precision better than 10 nm/s^2 is obtained after averaging data over a few minutes (Figure 3b; Carbone et al., 2019).

To examine the relation between the two gravity time series, we employ the wavelet coherence analysis (Grinsted et al., 2004). This method highlights how coherent the cross-wavelet transform of two time series is and can be thought as a localized correlation coefficient in the time-frequency space. The wavelet coherence plot (Figure 3c) indicates, over wavelet scales corresponding to periods longer than 4–5 days, high coherence during two subintervals: late August–mid-September (h-c 1) and mid-October–early November (h-c 2). In both cases, arrows pointing right indicate in-phase behavior. During the first subinterval, weak oscillations over time scales of about 5 days occurred around an almost constant level of the gravity signal (Figure 4a). During the second subinterval, an overall gravity decrease was observed (Figure 4b; the average rate of change of the signal from PDN is $-7.1 \text{ nm/s}^2/\text{day}$ during h-c 2, compared to an overall rate of $-1.6 \text{ nm/s}^2/\text{day}$). For the two subintervals of high coherence, the amplitude ratio between the signals from PDN and SLN is calculated as the slope of the best linear fit between the two time series (Poland & Carbone, 2016). Results point to PDN/SLN amplitude ratios of ~ 3 and ~ 6 , during the first and second subinterval, respectively (Figures 4c and 4d).

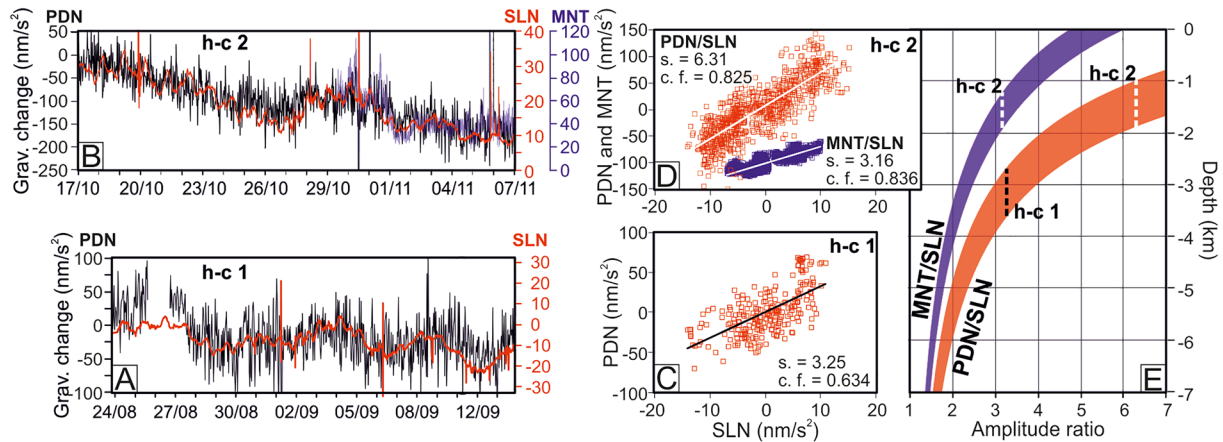


Figure 4. Gravity time series during high-coherence phases and derivation of mass source depths. (a and b) Gravity time series from PDN (black curves) and SLN (red curves), during the first (h-c 1) and second (h-c 2) subintervals of high coherence. As for h-c 2, the available signal from MNT is also shown (blue curve in panel (b)). (c and d) Scatterplots of SLN against PDN, during h-c 1, and SLN against PDN and SLN against MNT, during h-c 2. For each scatterplot, the slope of the best-fitting line (s.) and the correlation coefficient (c. f.) are reported. (e) PDN/SLN (red area) and MNT/SLN (blue area) gravity amplitude ratio versus depth of the mass source. The calculations are performed for all the possible positions of the mass source enclosed in the yellow area of Figure 1a. Dashed segments mark the values of the observed amplitude ratios during h-c 1 and h-c 2.

During the period under study, the other operating SG in the mini-array of Mt. Etna (La Montagnola hut; MNT; Figure 1a) worked only intermittently, due to problems in the data acquisition system. The available data cover only the last part of the h-c 2 interval and shows a decreasing pattern which is well correlated with the changes observed at PDN and SLN (Figure 4b). The MNT/SLN amplitude ratio during this interval is ~ 3 (Figure 4d).

5. Discussion

Only negligible ground deformation occurred during the two subintervals of high coherence (daily GPS data indicate changes within 0.2 and 0.5 cm, respectively, for the horizontal and vertical components), thus suggesting that the observed gravity changes are mostly due to subsurface mass redistributions. Furthermore, no important rain events occurred that could result in a $> 0.5 \text{ nm/s}^2$ change at SLN or PDN during the first 3.5 months of the period under study, which rules out the possibility that local hydrology may have somehow controlled these mass changes. Estimations of the depths of the gravity sources are made using the inversion method described by Carbone and Poland (2012) and reported in the Text S6 in Supporting Information S1. Assuming that the horizontal position of the gravity source lies within a ~ 2 -km-wide area enclosing the summit active craters of Etna (Figure 1a), each value of the gravity amplitude ratio will define a range of possible source depths, as depicted through the red (PDN/SLN) and blue (MNT/SLN) areas of Figure 4e. Accordingly, the ~ 3 PDN/SLN amplitude ratio during h-c 1 points to a depth of the mass source of between 2.5 and 3.5 km b.s.l. Conversely, the PDN/SLN (~ 6) and MNT/SLN (~ 3) amplitude ratios during h-c 2 consistently indicate a shallower mass source at 1–2 km b.s.l.

The couplings between the signal from the AQG-B and the signals acquired through SGs installed at sites 5 (MNT) and 9 (SLN) km away from PDN (Figure 1a) prove that, during the period under study, the AQG-B measured gravity changes reflective of bulk mass redistributions, rather than local and/or instrumental effects. Furthermore, the estimated source depths (Figure 4e) are in agreement with the results of previous studies (Carbone et al., 2003, 2019), thus suggesting that volcanic processes drove the observed gravity changes.

The gravity oscillations during h-c 1 are likely reflective of transient mass changes (on the order of $3 \cdot 10^{10} \text{ kg}$; see Text S6 in Supporting Information S1), possibly related to the pulsating ascent of gas-rich fluids toward the intermediate and shallow portions of the magmatic system (Paonita et al., 2021). The upward flushing of this fluid phase (Ferlito, 2018) is thought to control the occurrence and intensity of explosive activity at Mt. Etna (Moretti et al., 2018), implying that tracking it is important from the standpoint of volcano monitoring. Detection techniques based on geochemical measurements may fail to recognize the surface signature of this process, especially if it consists of minor gas emissions from the plume and from the flanks of the volcano, occurring over

relatively short time scales (e.g., a few days, as in the case of changes during h-c 1). In that sense, cross analysis with high-precision continuous gravity measurements provides a powerful tool to assess a more reliable picture of the volcano's state of activity.

The gravity decrease phase throughout h-c 2, corresponding to a mass decrease of about 6×10^{10} kg (Text S6 in Supporting Information S1), could indicate a gradual increase in the proportion of exsolved gas to magma in a shallower portion of the volcano's plumbing system. Early recognition of gas segregation processes at active volcanoes is of utmost importance, given the implications that local accumulations of volatiles may have for the development of energetic and potentially dangerous paroxysms (Allard et al., 2005; Carbone et al., 2008).

A decrease in the flux of SO_2 emitted by Mt. Etna's plume (from about 4,000 to about 2,000 tons/day) was observed during the first half of October (INGV-OE, 2020). Low degassing values persisted throughout the whole h-c 2 interval, while the SO_2 flux started increasing again since late-November. The phase of gravity decrease and low gas emission rate may have resulted from rheological stiffening in the upper conduits (Nadeau et al., 2011), inhibiting gas discharge and favoring accumulation at shallow depth. Cooling of the magma that fed mild Strombolian activity at two out of the four summit craters of Mt. Etna, during September and October 2020 (INGV-OE, 2020), could have led to the formation of the inferred stiffened layer. Also in this case, we show the importance of cross-checking geochemical observation with continuous gravity measurements: without the information from gravimetry, it is not possible to tell whether the observed decrease in the gas flux from the craters was due either to a lower supply from below, or to a reduced ability of the upper plumbing system to discharge the available gas.

6. Conclusive Remarks

With respect to instruments that have been previously utilized for continuous gravity measurements under harsh environmental conditions, the AQG-B has shown several advantages. It provides driftless measurements, allowing to precisely track changes over time-scales of days to months. Indeed, it would not have been possible to detect a gravity change like the one during the h-c 2 interval using a spring gravimeter, because of the marked nonlinear drift over periods longer than a few days (Carbone et al., 2017). Furthermore, thanks to the accuracy of the AQG-B, no fictitious changes in the level of the signal occur across gaps in the time series. Such "jumps" may occur when relative instruments are employed (spring meters and SGs), requiring arbitrary adjustments to compensate for changes in the baseline, that may eventually lead to wrong inferences about the driving processes.

Although the AQG-B features a lower short-term precision than the iGrav SG, it takes $\sim 1/3$ of the continuous power supply needed by that device, which allows for installation at sites with no mains electricity supply. The AQG-B is also easier to install and maintain than the iGrav, thanks to the lack of cryogenics and related equipment. Furthermore, inspection of the time series from the AQG-B at PDN did not reveal the occurrence of apparent changes resulting from the instrumental response to inertial forces, which were found in the signal from the iGravs at Mt. Etna, during phases of strong volcanic tremor (Carbone et al., 2019).

Field gravimetry is a relatively new area of application for quantum technology and there are ongoing improvements, both in terms of power consumption and of size and transportability of the instrumentation.

Cross analysis of the time series from the AQG-B and the iGravs at Mt. Etna revealed the occurrence of gravity changes likely driven by bulk volcanic processes. In particular, we propose that changes in the relative proportions of magma and exsolved gas in the upper part of the volcano's feeding system triggered the observed gravity changes.

Our successful demonstration of a quantum sensor based on atom interferometry opens new horizons not only for volcano gravimetry, but also for the application of the gravity method to contexts where reliable field measurements in hostile environments may be required, such as the study of groundwater dynamics, geothermal energy systems and carbon sequestration.

Conflict of Interest

B. Desruelle, V. M noret, L. Antoni-Micollier, and J. Lautier-Gaud declare the following competing interests: iXblue (formerly Muquans) is a privately held company. B. Desruelle, V. M noret, L. Antoni-Micollier, and J. Lautier-Gaud are employed by iXblue and hold shares in the company.

Data Availability Statement

The gravity data presented in this paper are available at: <https://zenodo.org/record/6668958#.YrAttXbPJ8> (doi: <https://doi.org/10.5281/zenodo.6668958>).

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