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# Continuous and near real-time measurements of gaseous elemental mercury (GEM) from an Unmanned Aerial Vehicle: A new approach to investigate the 3D distribution of GEM in the lower atmosphere

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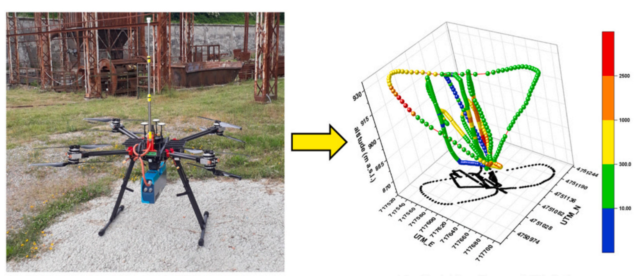
## HIGHLIGHTS

- First attempt to measure GEM in near real-time with a Lumex RA-915 M on an UAV.
- Test in selected sites of mining and urban zones of Abbadia San Salvatore (Italy).
- The method shed light on GEM spatial distribution and concentration variability.
- 3D dot-maps allowed to verify whether the guideline concentrations were exceeded.
- GEM contents achieved the highest concentrations above the old furnaces facilities.

## GRAPHICAL ABSTRACT



Lumex + UAV    3D distribution of GEM

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## ABSTRACT

We present the first real attempt to directly and continuously measure GEM through a Lumex RA-915 M, designed for real-time detection of mercury vapor, mounted on an UAV (Unmanned Aerial Vehicle, namely a heavy-lift octocopter), inside and outside the former Hg-mining area of Abbadia San Salvatore (Mt. Amiata, Italy), known as a GEM source. We tested the effectiveness of the UAV-Lumex combination at different heights in selected sites pertaining to both mining facilities and surrounding urban zones, shedding light on the GEM spatial distribution and concentration variability. The Lumex great sensitivity and the octocopter optimal versatility and maneuverability, both horizontally and vertically, allowed to depict the GEM distribution in the atmosphere up to 60 m above the ground. The acquisition system was further optimized by: i) synchronizing Lumex and UAV GPS data by means of a stand-alone GPS that was previously synchronized with Lumex; ii) using a vertical sampling tube (1.20 m high) connected to the Lumex inlet to overcome the rotors strong airflows and turbulence that would have affected GEM measurements; iii) supplying the octocopter with batteries for power supply to

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avoid the release of exhaust gases; iv) taking the advantage of the UAV ability to land in small spaces and stop at selected altitudes.

The resulting dot-map graphical representations, providing a realistic 3D picture of GEM vertical profiling during the flights in near real-time, were useful to verify whether the guideline concentrations indicated by competent authorities were exceeded. The results showed that the GEM concentrations in the urban area, located a few hundred meters from the mining structures, and close to already reclaimed areas remained at relatively low values. Contrarily, GEM contents showed significant variations and the highest concentrations above the facilities containing the old furnaces, where increasing GEM concentrations were recorded at decreasing heights or downwind.

## 1. Introduction

Air quality assessment and emission and distribution of pollutants in the atmosphere are a key issues of the European and global environmental policies, in order to both protect human health and environment from adverse effects by contaminants and develop concrete actions to support a modern ecological and green transition (WHO, 2016; EEA, 2020). Mercury is a toxic and noxious element for humans and ecosystems and gaseous elemental mercury (GEM or $Hg^0$) is considered a global and dangerous air pollutant due to its volatility and chemical inertia (WHO, 2000, 2007; Fitzgerald and Lamborg, 2007; Driscoll et al., 2013). The residence time of GEM in the atmosphere has indeed been estimated to be comprised between 6 and 24 months (Schroeder and Munthe, 1998; Lamborg et al., 2002). GEM is released by a variety of natural (e.g. Pyle and Mather, 2003; Engle et al., 2006; Bagnato et al., 2011, 2014; Tassi et al., 2016; Gagliano et al., 2016, 2019; Venturi et al., 2019; Edwards et al., 2020; Cabassi et al., 2021) and anthropogenic sources (e.g. Eckley and Branfireun, 2008; Pacyna et al., 2010; Pirrone et al., 2010; Huang et al., 2011; Streets et al., 2011; Acquavita et al., 2017; Sundseth et al., 2017; Tao et al., 2017; Cabassi et al., 2020). Contamination by mining and smelting activities where cinnabar (HgS)-rich ore deposits were converted to liquid mercury is known to be significantly high (e.g. Ferrara et al., 1998a; Fantozzi et al., 2013; Higueras et al., 2013, 2014; Barago et al., 2020; Esbrí et al., 2020; Floreani et al., 2020). Research and monitoring programs are presently aimed at carrying out inventory and controlling and minimizing the emissions from all relevant sources, enforcing the knowledge on the dispersion mechanisms of mercury in the atmosphere, as established by the UN Minamata Convention on Mercury, which came into force in August 2017 (UNEP, 2013). The Minamata Convention is currently implemented by dedicated actions, as those promoted by the Global Mercury Partnership (Bank et al., 2014) and its priorities for action (e.g. mercury air transport and fate research area; Mason and Pirrone, 2009). In this framework, both CNR-IGG and the Department of Earth Sciences of Florence actively participate as members. For this purpose, investigating and proposing new methods, approaches and/or protocols of measurement of gaseous mercury emissions become crucial for monitoring the presence of mercury pollution sources as well as their effects on the environment.

Unmanned Aerial Vehicles (UAVs), with their different shapes and sizes (Villa et al., 2016a,b; Hassanalian and Abdelkefi, 2017; Roch, 2020), currently represent the new frontier of monitoring environmental and air pollutants, since they can be used in many different contexts, ranging from major cities and urban regions (e.g. Gallacher, 2016a, 2016b; Lambey and Prasad, 2021 and references therein) to active degassing volcanoes and hydrothermal areas (Astuti et al., 2008; Stix et al., 2018; Liu et al., 2019; Mandon et al., 2019; Galle et al., 2021; Tamburello et al., 2021). UAVs are indeed suitable for different purposes (Burgués and Marco, 2020; Lambey and Prasad, 2021), such as field determination of: i) multi-pollutants (Aurell et al., 2017; Qiu et al., 2017), ii) sampling and/or measurements of specific gaseous species (McGonigle et al., 2008; Rossi et al., 2014; Chang et al., 2016; Rossi and Brunelli, 2017; Ruiz-Jimenez et al., 2019; Rutkauskas et al., 2019), or iii) detection of natural or anthropogenic pollutant plumes and modelling (Neumann et al., 2013; Barchyn et al., 2019). UAVs allow to mount devices for direct gas sampling or measurement in air and they are relatively low-cost and characterized by high sampling resolution, repeatability and flexibility and efficiency in 3D monitoring although they have so far limited flight time. Consequently, UAVs are preferable over other air quality monitoring platforms especially in dangerous or inaccessible environments (Villa et al., 2016a; Lambey and Prasad, 2021).

As far as gaseous mercury is concerned, a first application using a UAV was only recently made (Black et al., 2018), since continuous monitoring is generally performed at ground level and at fixed points (e.g. Wängberg et al., 2016) or by moving along pre-defined transects with portable devices (e.g. Vaselli et al., 2013; Cabassi et al., 2017), whereas measurements in the atmosphere are usually performed via airships (Slemr et al., 2009; Deeds et al., 2013). Black et al. (2018) collected gaseous mercury from anthropogenic emissions on gold-coated quartz cartridges allocated on a quadcopter, which was then analyzed via cold vapor atomic fluorescence spectrometry. To the best of our knowledge, direct, continuous and near real-time measurement are not so far reported by mounting a portable instrumentation for GEM determination housed in an UAV. Accordingly, this study was aimed at obtaining the very first measurements of GEM carried out up to an altitude of about 60 m above the ground by coupling a Lumex RA-915 M Mercury Analyzer, designed for real-time detection and monitoring of mercury vapor, to a heavy-lift octocopter. The GEM data were acquired via pre-established transects crosscutting the former Hg mining area of Abbadia San Salvatore (Mt. Amiata, central Italy), which is known to be an important GEM source (e.g. Bacci et al., 1994; Ferrara et al., 1998b; Vaselli et al., 2013; McLagan et al., 2019). The aim is to evaluate the 3D spatial distribution of GEM at different heights and testing the effectiveness of the UAV-Lumex combination. Consequently, UAV flights were carried out close to the mining facilities, while others were performed to verify whether Abbadia San Salvatore was impacted by the GEM released from the mining site, the urban areas being located a couple of hundred meters away.

## 2. The study area and the UAV flights

The study area is located inside the town of Abbadia San Salvatore (Fig. 1a and b), i.e. the most important site of cinnabar exploitation and mercury production of the Mt. Amiata Hg district (e.g. Vaselli et al., 2013 and references therein). Numerous studies (e.g. Gray et al., 2014; Rimondi et al., 2014, 2015, 2020; Vaselli et al., 2015, 2021; Magi et al., 2018; Lazzaroni et al., 2020; Pribil et al., 2020) highlighted that the Hg contamination and dispersion affected all the environmental compartments, i.e. geosphere, hydrosphere, pedosphere and atmosphere. According to the mining reports, at least 100,000 tons of liquid mercury were produced and about 10,000 tons of gaseous mercury were dispersed in the atmosphere (Ferrara et al., 1998b; Vaselli et al., 2017). The mining complex covers about 65 ha and hosts abandoned edifices, consisting of furnaces, driers, condensers, laboratories and technical and workers' buildings (Fig. 1b). Currently, remediation activities are in progress and aimed at achieving indoor and outdoor GEM concentrations of $<$500 and $<$ 300 ng $m^{-3}$, respectively (Regional Decree n° 1447, November 23, 1998; Vaselli et al., 2019). The most Hg-contaminated sites are associated with the edifices hosting the Gould and Nesa furnaces and condensers (up to >50,000 ng $m^{-3}$; Vaselli et al., 2017, 2019).

GEM anomalies were distributed outside the former mining area (Vaselli et al., 2013; McLagan et al., 2019). Consequently, the flights of the UAV-Lumex were planned to cover both the former mining site where the HgS-rich ore material was stored and liquid Hg was produced as well as the surrounding areas. In August 2020, six distinct flights, whose duration was of about <15 min each, were carried out and the takes offs were from:

- "Stadio": the local municipal stadium located inside Abbadia San Salvatore (Fig. 1c);
- "Altone": recreational and sport center in the western part of Abbadia San Salvatore (Fig. 1d);
- "Laghetto Verde": a small lake NW of the mining area (Fig. 1e);
- "Pozzo Garibaldi": Garibaldi mine shaft, positioned in the northern portion of the mining area, was used by the miners to reach the different levels (down to −400 m) in order to exploit the Hg-ore deposits, extract the material and manage and maintain the mining equipment (Fig. 1f);
- "Asciugatoi Vecchi": the old dryers situated in the central part of the mining area (Fig. 1g);
- "Forni": the edifices hosting the Gould and Nesa furnaces (Fig. 1h).

From each site, the UAV-Lumex was travelling along by following predetermined routes.

## 3. Materials and methods

The UAV selected for the atmospheric surveys is a Hammer X8B, a heavy-lift coaxial octocopter originally designed for the film industry (Fig. 2a, b, c). Its versatility makes it a suitable choice for such activities since a high degree of precision as well as operation safety during overflight are required. The X8B can carry payloads up to 20 kg. It has a full carbon fiber airframe measuring 140 cm diagonally (motor to motor), which consists of a central rigid core structure, where flight controller, batteries and payload are located, and four folding arms where the motors are mounted (Fig. 2b). The aircraft eight propulsion units (each consisting of a motor and propeller) are arranged in coaxial X8 configuration with four arms carrying two contra-rotating units each. The use of multiple motors allows the UAV to maintain stable fight even in the unlikely event of a motor failure. While it is less efficient than a "flat" (not coaxial) layout, a notable advantage of the X8 design, when compared to a flat 8, is that with 4 as opposed to 8 arms, larger motors and propellers can be fitted for increased lifting capacity without greatly increasing the size of the aircraft. A further benefit of a coaxial layout is that it greatly reduces exposed propeller disk area when compared to an equivalent flat motor layout, making the aircraft far less sensitive to wind conditions. The motors are powered by four 14,000 mAh 6S (22.2V) Lithium Polymer (LiPo) batteries arranged in two separate 14,000 mAh 12S (44.4V) banks joined in parallel for a total of 28,000 mAh capacity. The setup ensures the aircraft to have enough reserve power to make a safe landing even in the event of battery malfunctioning. The aircraft is controlled via a DJI A3 Pro flight controller and a DJI Lightbridge 2 control link. The A3 Pro is a triple redundancy flight controller with three GPS units, three magnetometers, and three IMUs working together, so that a reliable backup is always available in the event of a failure. The Lightbridge 2 Control Link allows the use of a ground station running the DJI GO app, which provides full real-time telemetry data for aircraft position, altitude and speed, as well as acting as a flight data recorder. The use of real-time positioning data with satellite map overlay allows precise positioning of the aircraft over the areas to be investigated.


**Fig. 1.** Location of Abbadia San Salvatore (Mt. Amiata, central Italy) (a) and the former Hg mining area (b). The flight routes carried out by the UAV-Lumex pair at the selected six sites are also reported: "Stadio" (c), "Altone" (d), "Laghetto Verde" (e), "Pozzo Garibaldi" (f), "Asciugatoi Vecchi" (g) and "Forni" (h). See text for additional information.

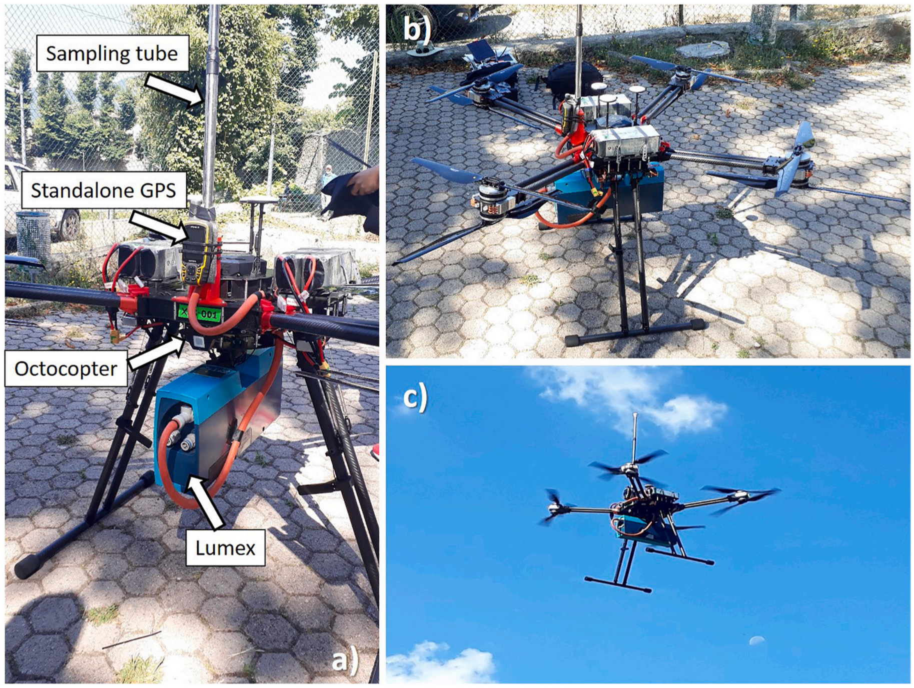


**Fig. 2.** The Hammer X8B heavy-lift coaxial octocopter coupled with Lumex that was equipped with a sampling tube and stand-alone GPS on-board (a–b). UAV-Lumex pair during the flight (c). See the text for further details.

The main technical UAS (Unmanned Aircraft System, i.e. an unmanned aircraft and the equipment to control it remotely; EU, 2019) characteristics are, as follows:

- Airframe: Hammer X8B
- Motors: 8 T-Motor MN 701-S
- Motor Controllers: 8 T-Motor ALPHA 60A HV
- Propellers: 8 T-Motor 26.2X8.5
- Batteries: 4 Tattu 14000 6s (22.2v) 24C
- Flight Controller: DJI A3 PRO
- Control Link: DJI Lightbridge 2
- Ground Station: iPad Pro running DJI GO

The Lumex RA-915 M Mercury Analyzer is based on differential atomic absorption spectrometry using high-frequency modulation of light polarization (ZAAS-HFM) (Sholupov et al., 2004) and is able to continuously measure GEM concentrations (range: 2:50,000 ng m$^{-3}$) in real-time and at high frequency (up to 1 s, as in the case of this work). The accuracy of the method is 20% (Sholupov and Ganeyev, 1995). A zero correction system continuously checks the baseline during sampling, while a zero check of about 40 s is performed at the beginning and at the end of each measurement session by using an internal calibration cell. The Hg device was fitted underneath the UAV (Fig. 2a) and custom designed parts, 3D printed in PLA, were used to mount the sampling tube in a vertical configuration 1.2 m above the body of the aircraft. This installation was chosen to minimize the potential risk of false readings due to sampling air disturbance by the aircraft. Additionally, a stand-alone GPS unit (Fig. 2a), synchronized with the Lumex at the same data acquisition frequency, was mounted on the aircraft as a yardstick to temporally compare the Lumex measurements and provide a backup for the GPS data from the Ground Station.

Great care was taken during the flights to minimize as much as possible the risk of producing unreliable measurements in the case the UAV-Lumex pair was overlapping air portions where the unmanned vehicle was previously passed and turbulence developed. This was achieved by controlling the UAV in a way that it was flying diagonally, i.e. oppositely to the vertical ascent and descent profiles. The measurements were performed every 10 m increments between 10 m and 60 m above ground level, in some cases spending a minimum of 30 s at each altitude to ensure representativeness, or by flying an ascent/descent never exceeding 1 m s$^{-1}$ and 5 m s$^{-1}$ vertical and horizontal speed, respectively. In some cases, the GEM data were acquired at fixed altitudes while moving across the areas of interest. When the measurement flights were concluded, the flight data from the Ground Station were coupled with those from the Lumex and the standalone GPS.

In the Supplementary Material, two videos show the UAV-Lumex pair flying over the edifices hosting the Gould and Nesa furnaces ("Forni" site, Video 1) and the soft landing of the UAV-Lumex pair after measuring GEM at "Altone" site (Video 2).

Supplementary video related to this article can be found at https://doi.org/10.1016/j.chemosphere.2021.132547

## 4. Results

The number of measurements (n) and the GEM data of the six surveys are summarized in Table 1, along with the geographical coordinates of the sites and the data of altitude (in m) and flight time (CET: Central European Time) registered by the UAS. The meteorological parameters (temperature in °C, wind direction in °, wind speed in m sec$^{-1}$ and humidity in %) refer to the local ground weather recorded by a weather website that interfaces with the flights records. All data can be accessed directly from the drone management portal owned by ©Airdata UAV, Inc. During the flights, wind direction was between 58 and 75° (approximately from ENE) with a wind speed up to 1.8 m s$^{-1}$, while air temperature and humidity ranged from 27.9 to 31.1 °C and from 20 to 30%, respectively, in relation with the flight time. The minimum altitude varied from 828 m ("Stadio") to 897 m ("Laghetto Verde" and "Pozzo Garibaldi"), the maximum from 883 m ("Stadio") to 962 m ("Laghetto Verde"), whilst the mean altitude value varied between 865 and 933 m. The GEM concentrations during the flights from "Stadio", "Altone", "Laghetto Verde" and "Pozzo Garibaldi" did not show a significant variability, being comprised between 18 and 33 ng m$^{-3}$ and having almost coincident geometric average, mean and median values (Table 1). On the contrary, with regard to the flights over and around the

**Table 1**
Geographic coordinates (UTM WGS84), number of measurements (n), minimum, maximum, geometric average, mean, median and standard deviation values of GEM (ng m$^{-3}$), minimum, maximum and mean values of altitude (m), temperature (°C), wind direction (°) and speed (m s$^{-1}$), humidity (%) and flight time (hh:mm) for the six selected sites. The flights date is August 10, 2020. See the text for further details.

| Site | "Stadio" | "Altone" | "Laghetto Verde" | "Pozzo Garibaldi" | "Asciugatoi Vecchi" | "Forni" |
|---|---|---|---|---|---|---|
| UTM_E | 718352 | 717734 | 717405 | 717401 | 717568 | 717625 |
| UTM_N | 4751051 | 4750799 | 4751226 | 4751393 | 4751158 | 4751104 |
| n | 268 | 510 | 457 | 290 | 116 | 725 |
| Min. GEM | 23 | 19 | 21 | 18 | 8.5 | 5.2 |
| Max. GEM | 30 | 30 | 33 | 30 | 696 | 5009 |
| Geom. Aver. GEM | 27 | 26 | 24 | 20 | 81 | 120 |
| Mean GEM | 27 | 26 | 25 | 20 | 135 | 491 |
| Median GEM | 28 | 26 | 24 | 20 | 101 | 105 |
| St. Dev. GEM | 1.3 | 2.0 | 2.1 | 1.6 | 141 | 843 |
| Min. Altitude | 828 | 870 | 897 | 897 | 881 | 873 |
| Max. Altitude | 883 | 924 | 962 | 950 | 931 | 934 |
| Mean Altitude | 865 | 902 | 933 | 926 | 899 | 902 |
| Temperature | 27.9 | 28.7 | 29.8 | 30.4 | 30.8 | 31.1 |
| Wind direction | 75 | 71 | 66 | 63 | 61 | 58 |
| Wind speed | 1.8 | 1.8 | 1.8 | 1.7 | 1.6 | 1.6 |
| Humidity | 30 | 27 | 24 | 22 | 21 | 20 |
| Flight time | 10:40 : 10:45 | 11:14 : 11:23 | 12:02 : 12:10 | 12:30 : 12:35 | 12:52 : 13:00 | 13:16 : 13:29 |

"Asciugatoi Vecchi" and "Forni", the GEM concentrations varied between 8.5 and 696 ng m$^{-3}$ and between 5.2 and 5009 ng m$^{-3}$, respectively, thus exceeding as expected the outdoor limit value recommended by local regulations (300 ng m$^{-3}$; Regional Decree n° 1447, November 23, 1998; Vaselli et al., 2019) and, for the "Forni" site, the guideline for inorganic mercury vapor of 1000 ng m$^{-3}$ as an annual average (WHO, 2000). Accordingly, their median values (101 and 105 ng m$^{-3}$, respectively) did not coincide with their geometric averages (81 and 120 ng m$^{-3}$, respectively). The GEM standard deviation values were from 1.3 ("Stadio") to 843 ("Forni") ng m$^{-3}$ (Table 1). The data distribution of each site is shown in the box-plot charts of Fig. 3.

## 5. Discussion

Spatial atmospheric measurements are becoming essential for air pollution forecasting and monitoring (Gu et al., 2018), particularly in environments characterized by contaminants with relatively high volatility and residence time in the atmosphere such as GEM. Fixed platforms for high frequency continuous measuring cannot correctly account for the spatial variability of gases (Gu et al., 2018). On the other hand, low-cost passive traps or samplers placed in multiple sites within a selected area are regarded as useful to understand the distribution of pollutants (e.g. McLagan et al., 2016, 2019). However, they cannot be able to recognized sudden or short-time events of contamination since they are exposed for relatively long-time (e.g. weeks) and consequently, they retrieve weighted average concentrations (e.g. Venturi et al., 2016). Differently, UAVs combined with instrumentation or sensors for continuous measurements are the correct approach for the growing need to understand the 3D spatial distribution of atmospheric pollutants. They are indeed able to provide the contaminant near-surface vertical profiling over large or site-specific areas in near real-time (Gu et al., 2018; Rutkauskas et al., 2019; Burgués and Marco, 2020; Lambey and Prasad, 2021). The UAV-Lumex pair used in this work allowed to acquire a 3D spatial distribution of GEM in the former Hg-mining facilities of Abbadia San Salvatore and surroundings, being able to instantaneously catch the presence of local anomalies and, moreover, when present, to follow and evaluate the Hg-dispersion halo.

The approach applied during the UAV flights requires the correct synchronization of Lumex and UAV GPS data since the internal clocks of the two devices are working independently, being separate components (Gu et al., 2018). This issue was bypassed by synchronizing the stand-alone GPS and the Lumex. Consequently, each single value had its own corresponding georeferencing data. In Fig. 4 (a, b), an example ("Forni" site) of the acquired GPS data is reported and compared with those of the UAV, demonstrating that the two acquisitions overlap perfectly, i.e. the GEM data were synchronous with the UAV position data. During the monitoring surveys by rotary-wing vehicles, the rotors generate very strong airflows (Burgués and Marco, 2020) that affect the

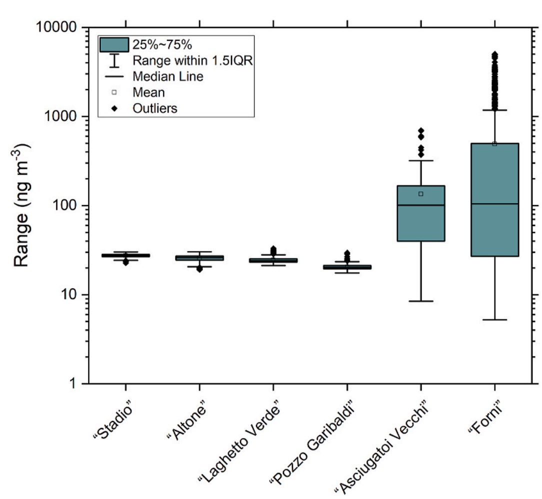


**Fig. 3.** Box-plot chart of the measured GEM concentrations (in ng m$^{-3}$) in the selected six sites.

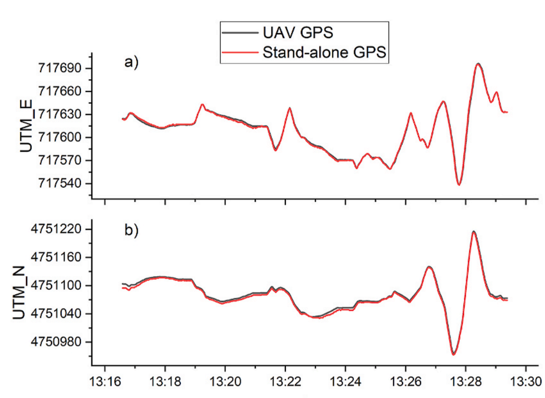


**Fig. 4.** UAV (black lines) and stand-alone GPS (red lines) data at "Forni" site for UTM_E (a) and UTM_N (b) coordinates. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

air distribution around the UAV and consequently, the data acquired by the on-board sensor. This drawback was overcome through a vertical sampling tube connected to the Lumex inlet. Such a configuration, requiring the measurement to be carried out at 1.2 m above the aircraft, allows to avoid the so-called "downwash", i.e. rotors vertical airflow affecting the air masses for several meters below the UAV and having worst effects especially across strong spatial gradients (Burgués and Marco, 2020 and references therein). Other important advantages of the UAV chosen for this study are: 1) the use of batteries for power supply, which avoid the release of exhaust gases from the engine (Black et al., 2018) and 2) the drone ability to land in small spaces and stop at pre-defined altitudes, which proved to be fundamental in our case, being very close to the emission points of the former mining area (Burgués and Marco, 2020).

The GEM dispersion was visualized by using dot-map graphics, i.e. a map based on selected concentrations intervals plotted with different colored circles (e.g. Cabassi et al., 2017). This simple representation allows to depict the GEM variation along the UAV flight transects. As shown in Fig. 5, the concentrations during the flights at "Stadio" (Fig. 5a), "Altone" (Fig. 5b), "Laghetto Verde" (Fig. 5c) and "Pozzo Garibaldi" (Fig. 5d) sites maintained at relatively low values and did not substantially change both horizontally and vertically. This result corresponds to the site-specific features, since the first two sites are located a few hundred meters from the former-mining area, while the second two sites have already undergone considerable remediation work, respectively. This also testifies that during the measurements the urban area of Abbadia San Salvatore was not affected by significantly high GEM concentrations although the measured values were recorded to be higher than those of background values for Mt. Amiata (3–5 ng $m^{-3}$; Ferrara et al., 1998b). On the other hand, the "Asciugatoi Vecchi" and "Forni" flights registered a large variability in terms of GEM concentrations, although in the first case a few minutes of incorrect Lumex data acquisition with respect to the zero check occurred, preventing a correct graphical representation of the mercury contents. In Fig. 6, the "Forni" dot-map highlights the GEM dispersion plume originated by the mining facilities mostly released from the edifices where the Gould and Nesa furnaces are hosted. In fact, the GEM concentrations were higher than those required for outdoor value (300 ng $m^{-3}$) and those reported in the guidelines for inorganic mercury vapor (1000 ng $m^{-3}$; WHO, 2000). In this area, as well as around the "Asciugatoi Vecchi" site, Vaselli et al. (2013, 2017) measured, at 150 cm from the ground, GEM contents between 5000 and 10,000 ng $m^{-3}$ and >10,000 ng $m^{-3}$ and related to the presence of calcines, Hg-rich untreated material and liquid Hg. The highest concentrations (even >2500 ng $m^{-3}$) measured by the UAV-Lumex pair were found (Fig. 6), as follows: 1) in close proximity to the ground, concordantly with an increase with decreasing height (e.g. McLagan et al., 2019, 2021) and 2) in the southeastern portion of the air route and at 30–40 m altitude, i.e. consistently with an upward migration of GEM along the wind direction (Table 1). Actually, air movement and spatial variation of gaseous mercury in a mining–metallurgical environment or in any contaminated site with multiple emissions and with major sources acting as a constant GEM supplier can principally be explained by dilution processes, as already pointed out by Esbrí et al. (2020) and McLagan et al. (2021). Furthermore, the choice to fly following diagonal trajectories, as well as to carry out measurements at fixed altitudes while moving across areas of interest, proved to be appropriated, allowing to identify large and sudden vertical GEM variations even at minimum distances. It is indeed worth noting that path planning and track design are pivotal for the successful completion of the flights and suitable environmental pollution detection by UAVs (De Filippis et al., 2012; Rohi et al., 2020).

## 6. Conclusions

The present study demonstrated for the very first time that direct, continuous and near real-time GEM measurements aboard an UAV allowed to recognize the air concentration variability between different sites and to identify sudden mercury changes around the emission sources. This was guaranteed by: i) the great sensitivity of the UAV-Lumex pair combination and the optimal versatility and maneuverability in both horizontal and vertical dimensions, ii) the high degree of precision of the heavy-lift octocopter, as well as iii) Lumex and UAV GPS

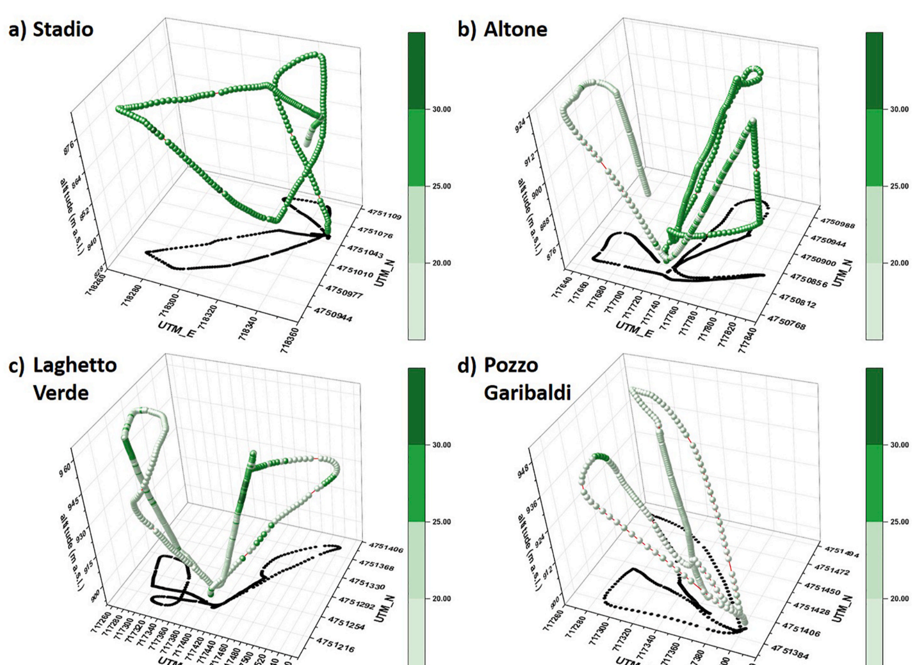


**Fig. 5.** 3D dot-maps of the GEM (in ng $m^{-3}$) measurements in air performed during the flights at "Stadio" (a), "Altone" (b), "Laghetto Verde" (c) and "Pozzo Garibaldi" (d) sites. A green color scale was used to depict the GEM concentrations. The black curves are the projection of the flight at the ground level. See the text for further details. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

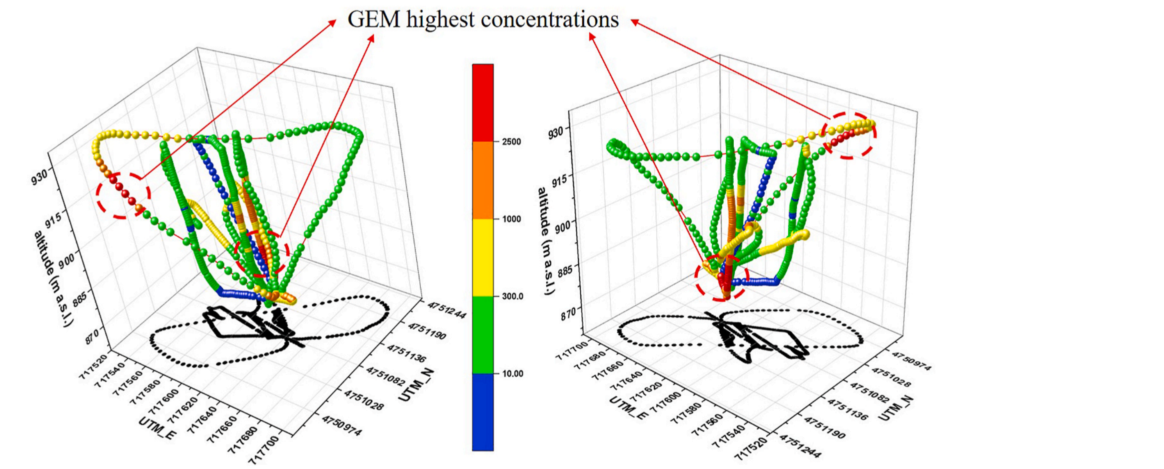


**Fig. 6.** 3D dot-map of the GEM (in ng m$^{-3}$) measurements in air performed during the flight at "Forni" site from two different perspectives. The highest measured concentrations are indicated. The black curves are the projection of the flight at the ground level. See the text for further details.

synchronization and iv) the use of a vertical tube for air sampling and batteries for power supply. The resulting dot-map graphical representation provided a realistic 3D picture of GEM distribution during the UAV flight, able to verify whether guideline concentrations indicated by international, national or local authorities were exceeded. This kind of measurements is in fact becoming necessary, especially close to areas known to be contaminated and placed next to inhabited centers, as in the case of Abbadia San Salvatore and the former mining area, in order to undertake appropriate actions to mitigate the possible risk for the local community and to successfully implement the Minamata Convention by means of emerging investigation techniques for GEM atmospheric monitoring (Gustin et al., 2016). In particular, the results showed that the GEM concentrations in the urban area, as opposed to those near the old furnaces buildings, remained roughly stable at relatively low values during the flights.

The presented instrumental approach is therefore able to overcome the limitations of both fixed measuring stations, which cannot take into account the GEM spatial variability, and passive samplers, which do not provide indications on concentration variations in short periods of time. The UAV-Lumex pair can further be improved and implemented by repeating the flights at different hours and days to account for the temporal evolution of the GEM three-dimensional spatial distribution, as shown by e.g. Vaselli et al. (2013), Esbrí et al. (2020) and McLagan et al. (2021) who recorded significant GEM variations between day and night. Additionally, the atmospheric conditions play a key role, as they are able to increase or decrease the GEM concentrations and their distribution. Furthermore, the UAS data acquisition accuracy and the experience of the UAV pilot can allow to standardize the flight operations, i.e. accurately repeating the flight paths in different environmental conditions since there is the possibility of reprogramming the route based on previously acquired coordinates and altitudes. Finally, the expenses are relatively limited, since once the UAV and Lumex are purchased the costs can be easily amortized, i.e. only the maintenance of the two instruments is required.

## Author contributions

**Jacopo Cabassi**: Conceptualization; Data curation; Formal analysis; Investigation; Methodology; Validation; Visualization; Writing - original draft; Writing - review & editing. **Marta Lazzaroni**: Formal analysis; Investigation; Methodology; Validation; Visualization; Writing - review & editing. **Luciano Giannini**: Conceptualization; Data curation; Formal analysis; Validation. **David Mariottini**: Conceptualization; Data curation; Formal analysis; Resources; Software; Validation. **Barbara Nisi**: Data curation; Formal analysis; Investigation; Visualization; Writing - review & editing. **Daniele Rappuoli**: Conceptualization; Funding acquisition; Methodology; Project administration; Resources; Supervision; Validation. **Orlando Vaselli**: Conceptualization; Funding acquisition; Investigation; Methodology; Project administration; Resources; Supervision; Writing - original draft; Writing - review & editing. All authors have read and agreed to the published version of the manuscript.

## Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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