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Utilization of *in situ* airborne MS-based instrumentation for the study of gaseous emissions at active volcanoes

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ABSTRACT

A small, 24 V powered, portable mass spectrometer system, named ULISSES, for the study and visualization of gaseous volcanic emission is described. First deployments of the system have focused on both ground and airborne *in situ* measurement to monitor the awakening of the Turrialba Volcano in Costa Rica. Key gas measurements were acquired prior and after its eruption on 5 January 2010, confirming the presence of gas chemistry precursors typical of volcanic eruptions. Ground and airborne measurements were acquired to gain volcanological insight and as the first step towards the use of unmanned aerial vehicles (UAVs) as future airborne platforms and to confirm its unique capability to serve as a calibration/validation tool for satellite remote sensing data. Low parts per million (ppm) levels of helium and a large concentration of sulfur dioxide were measured *in situ* after the initial eruption. In particular, the SO_2 data correlated with satellite remote sensing data.

1. Introduction

Part of the strategic plan for NASA's Earth Surface and Interior Focus Area [1] identifies a significant scientific challenge: "How do magmatic systems evolve and under what conditions do volcanoes erupt? For volcanology, one of the key science questions driving remote sensing instrumentation is: "Do volcanoes signal their impending eruptions through changes in surface temperature or gas emission rates, and are such changes unique to specific types of eruptions?"

To begin to answer these questions, researchers have demonstrated that changes in volcanic gas emission in some cases provide early warning, in the range of hours to days, in advance of hazardous volcanic activity [2–6]. Sampling of these indicator gases, however, must often be performed in close proximity to the fumarolic vents, thus placing people in harm's way. Alternately, several researchers have deployed sensor networks [3] to monitor increasing volcanic activity, but often new activity will manifest itself some distance from the point of a previous eruption, making such sensor nets expensive, inflexible and in some cases non-useful. Satellite remote sensing is an alternative that can often be useful [7–12], but spatial resolution, spectral calibration and cloud coverage offer challenges. An airborne multi-gas sensor that can be flown at low altitude to obtain a three-dimensional chemical map of a volcanic plume, as well as to provide suitable calibration and validation data for satellite-based remote sensing instruments addresses these issues.

The benefits of using mass spectrometry for *in situ* volcanic monitoring have been discussed in previous papers [6,13]. It offers multiple determinations of chemical species, together with a large dynamic range of concentrations—from parts per billion (ppb) to 100% concentration. It can perform both qualitative and quantitative analysis of any gas-phase species present, with sufficient sensitivity to detect even very minor components, making this analytical method a good candidate to achieve the ideal of a continuous volcanic gas-monitoring sensor.

Over the past decade, the Gas Sensing Lab at the University of Costa Rica has been working in collaboration with gas geochemistry researchers and volcanologists to provide complementary gas geochemical data to help monitor and understand

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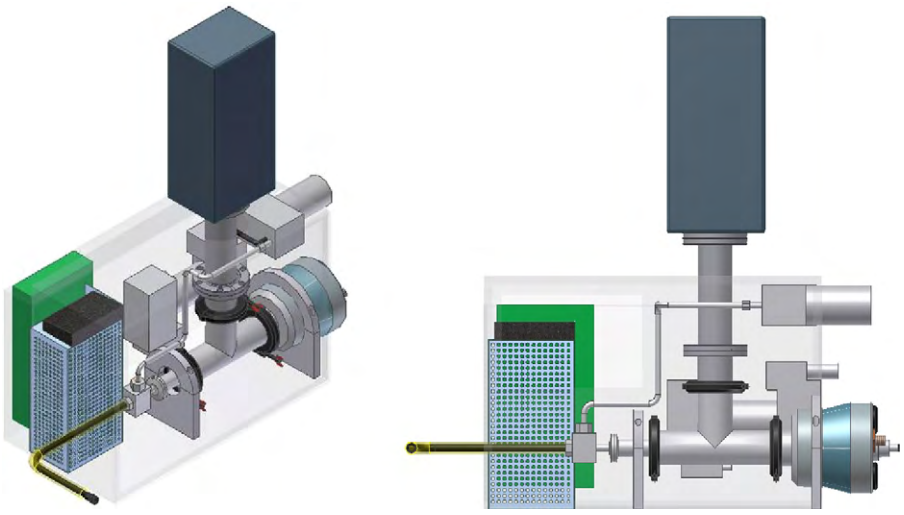

Fig. 1. ULISSES prototype with compact turbo-pump (ATH30+), two miniature diagram pumps (KNF 84.4 and 84.3), a small commercial mass analyzer (RGA 100 from SRS), control electronics and two 12 V batteries.

volcanic activity. Parallel to this applied research effort, the Hazard and Gas Detection Laboratory at NASA Kennedy Space Center has developed several small mass spectrometer systems [14,15]. These include an *in situ* mass spectrometer-based gas analyzer [16] that measures many volcano-activity marker species (VAMS). The instrument, named AVEMS for Airborne Volcanic Emissions Mass Spectrometer, was deployed and flight tested over active volcanoes, from 2003 to 2006 onboard the NASA WB-57 high-altitude research aircraft and Cessna 206 Stationair airplane providing the first *in situ*, NIST-traceable, three-dimensional, quantitative chemical plot of VAMS from several volcanoes in Costa Rica [17].

Some limitations of the current AVEMS instrument are its size (82,000 cm^3) and weight (32 kg), affecting its portability, and

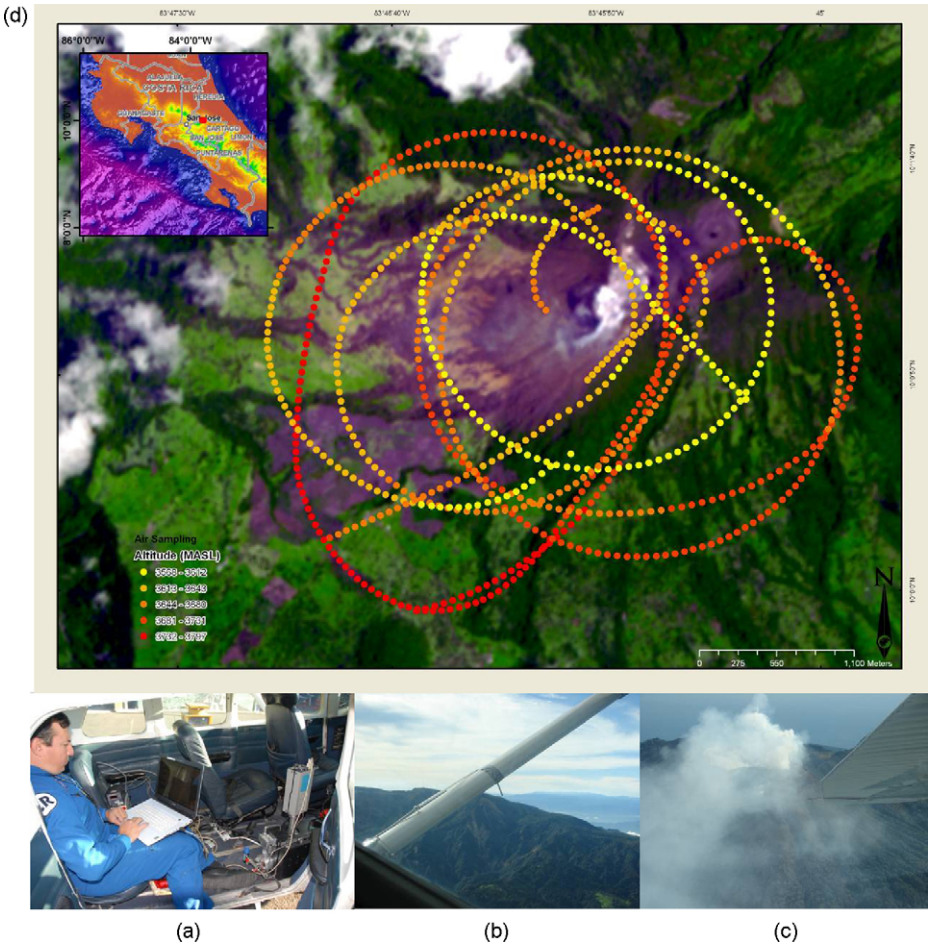

Fig. 2. ULISSES flight testing for *in situ* MS volcanic plume analysis. (a) Airborne MS and operator in pre-flight testing on CESSNA 206 aircraft. (b) Sampling port on wing. (c) Sampling Turrialba Volcano plume from aircraft. (d) GPS flight path and site geo-location for Turrialba Volcano.

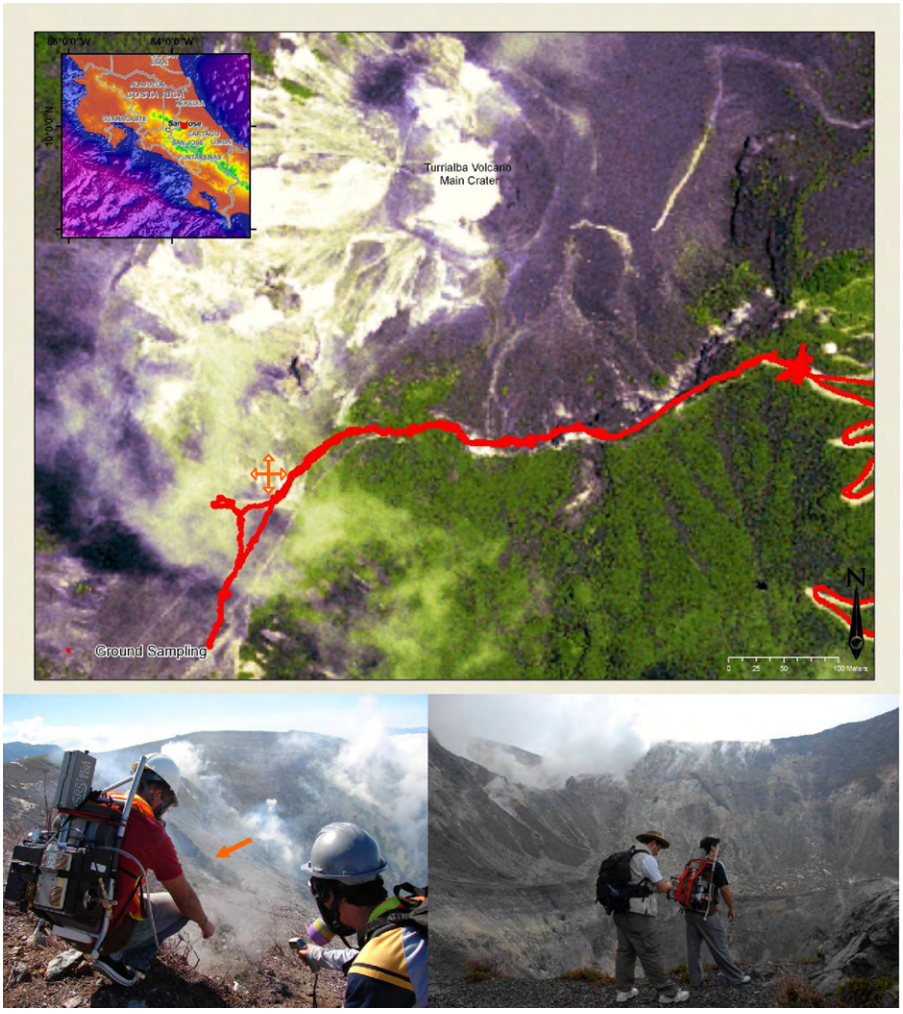

Fig. 3. ULISSES ground testing at Turrialba Volcano, system is transported as backpack by operator along with GPS unit tracking the route (in red) and sampling sites. The orange arrows mark the vent crated on the 5 January 2010 eruption.

thus its utility by a single person for ground sampling operations around volcanoes, for which eruption activity might be imminent. Its current size also necessitates a relatively large manned airborne platform, which restricts its usage in active volcanic eruption conditions when there is a release of hazardous ash clouds.

Described here is a new initiative to study and visualize gaseous volcanic emissions using mass spectrometer instrumentation. A

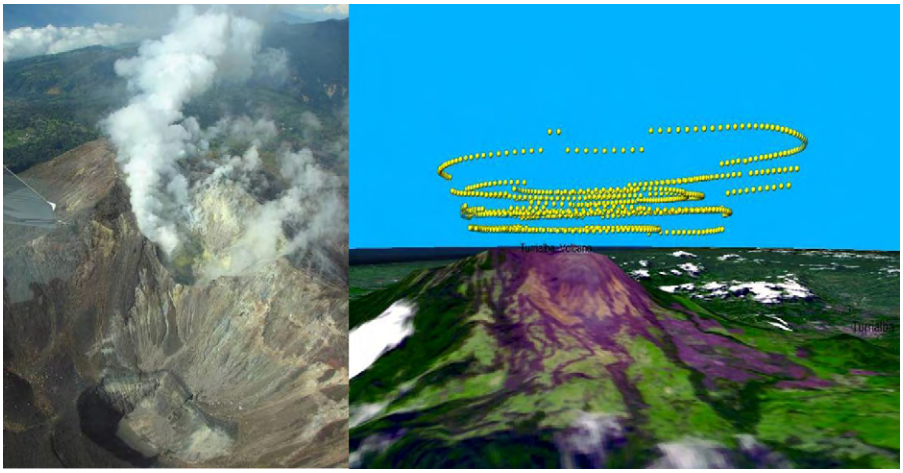

Fig. 4. Left: Turrialba Volcano September 2009 fly-over. Pre-eruptive conditions, active plume with high H_2O and CO_2 contents. Right: ULISSES 3D flight path visualization flown on CESNA 206 combined with 9 September 2009 ASTER satellite natural color composite image (BANDS Visible Near Infrared: 1, 3N, 2 and ASTER digital elevation model. Purple colors show damaged vegetation from the increasing temperature and toxic gases emission previous to the January 2010 eruption.

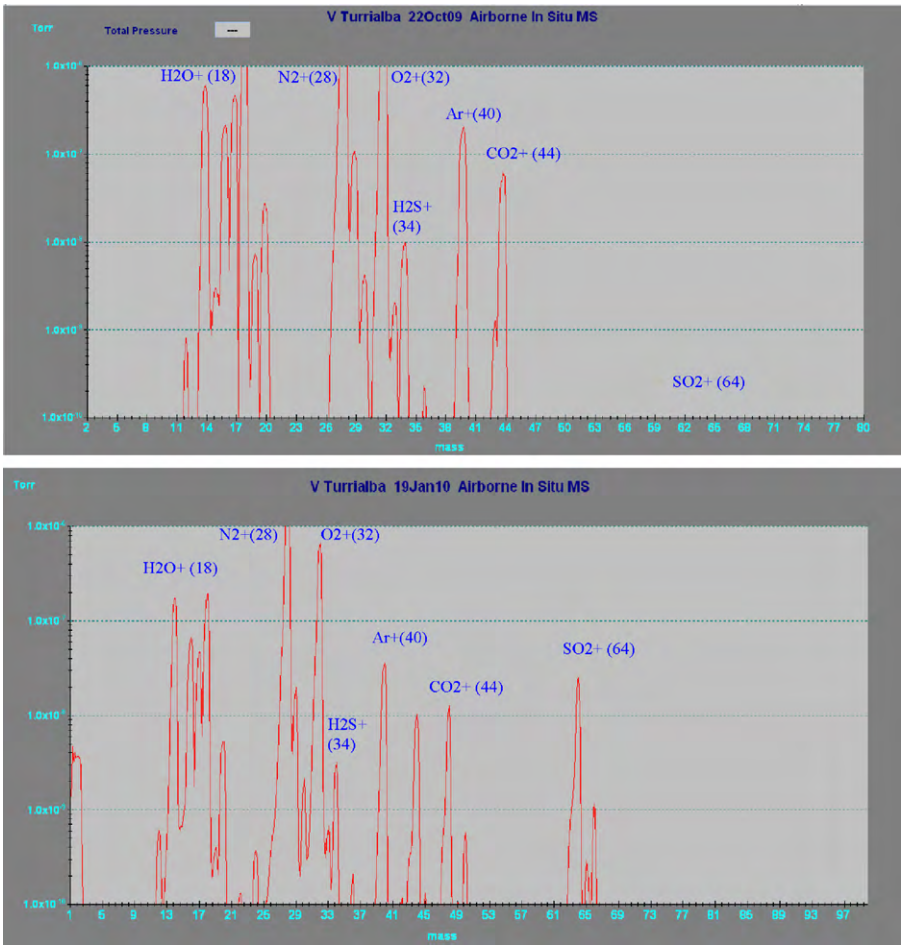

Fig. 5. ULISSES airborne data, full spectrum before (top) and after (bottom) the January 5th eruption showing air and water vapor peaks plus some of the principal VAMS components: SO_2, H_2S, CO_2. Carbon dioxide diminished while SO_2 was predominant after the eruption.

small *in situ* MS-based system named ULISSES (Utilization of Lightweight *in situ* Sensors and remote Sensing to study active volcanic Emissions Sites) was developed to demonstrate a path for single person use and for its integration into small unmanned aircraft vehicles (UAV), targeted to correlate *in situ* ground data to remote sensing satellite base data for calibration and validation purposes.

To further these efforts, three NASA centers – the NASA Ames Research Center (ARC), NASA Wallops Flight Facility (WFF), and the Jet Propulsion Laboratory (JPL) – are collaborating to establish a focus group on using small airborne UAV platforms, MS instrumentation, complementary chemical sensors and meteorological parameters to create an integrated single low altitude, harsh environment, system for *in situ* volcanic plume gas analysis.

2. Experimental/methods

The prototype ULISSES instrument is based on two previously developed and deployed systems—the Portable Double Focusing Mass Spectrometer system, used for *in situ* ground volcanic measurement [13,22], and the AVEMS instrument flown in the WB57 aircraft [16,17]. ULISSES is smaller than AVEMS with only a quarter of its size (21,000 cm^3) and a third of its weight (10 kg) but without auto-calibration or autonomous operation capabilities.

The RGA-100® from Stanford Research Systems Inc. is used as the analyzer to measure species with molecular weights up to 100 amu (Fig. 1). The decision to use this analyzer was based on several factors: first, significant past experience using the RGA-100 mass spectrometer on similar applications including volcano deployments in Kilauea Volcano. Second, the cost issue: Its one of the least expensive analyzers on the market, there are other high performance mass spectrometer but at higher prices, the target is to have a MS that could be damaged or lost on a volcano deployment. Finally, the results from a mass analyzer evaluation study that the HGDL performed during 2000–2001 and 2007–2008 periods [15,21] showed very good performance of the RGA for different harsh-environment gas-monitoring applications.

The RGA performs either in full spectrum mode or single ion monitoring. MS head parameters are 1 mA filament current, 70 eV electron acceleration, 90 eV focus acceleration, ion energy set to high, 1000 V electron multiplier voltage, 10 points per amu, 1–100 amu spectrum.

The MS system operates in the mid-10^{-5}-Torr pressure range. An Alcatel ATH® 30+ turbomolecular-drag high-vacuum pump is used to achieve the required vacuum. This pump was selected due to its high compression ratio, need for good response and recovery times between samples. Two diaphragm pumps are used in the system: a KNF 84.4 for roughing and a KNF 84.3 for inlet sample transport.

Sample is introduced to the mass spectrometer using a heated (150 cc at 1 atm differential pressure) molecular frit. The system requires 120 W of start-up power and 60 W at steady state. Power is supplied by two 12 V batteries, resulting in 3–4 h of operation. The system is manually activated and the MS is controlled by the operator via RS-232 communication using a small laptop computer.

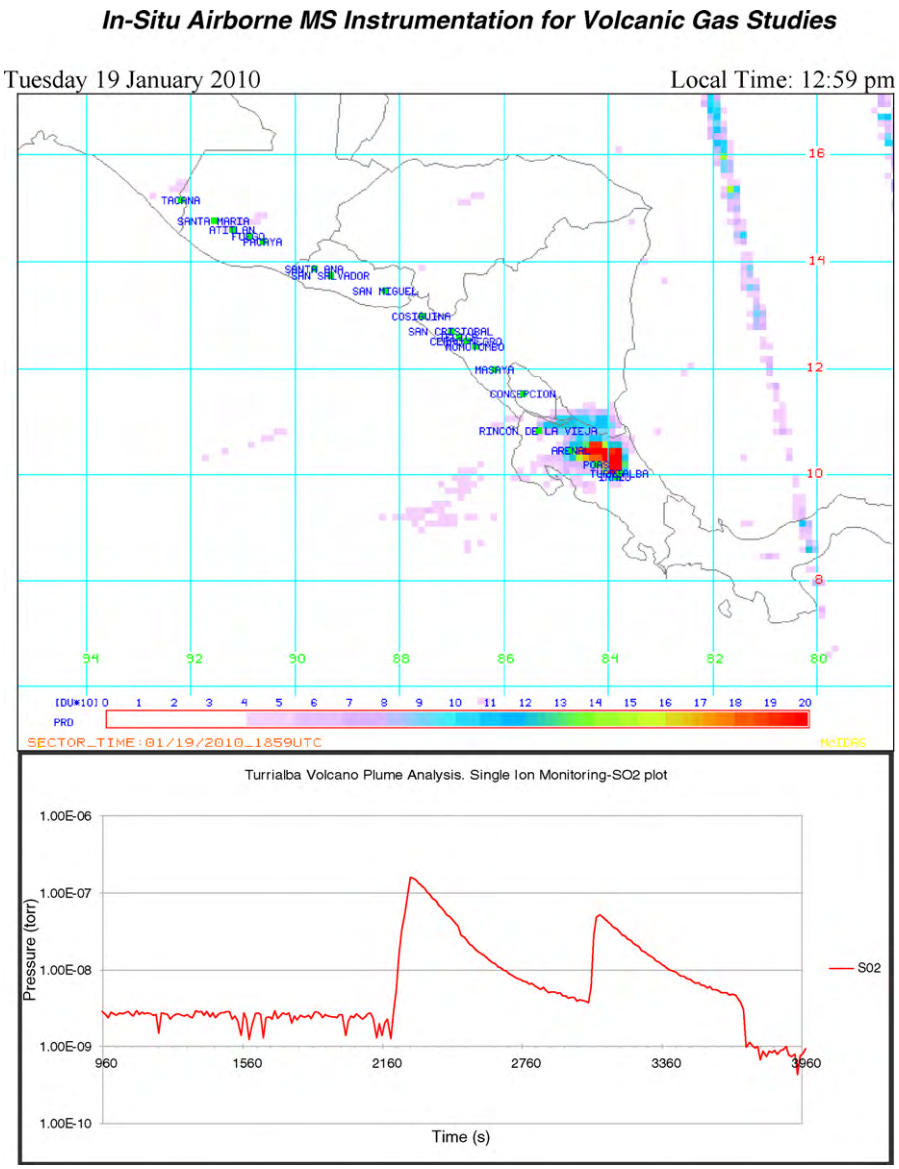

Fig. 6. ULISSES MS airborne data through the plume and correlation with +OMI_SO_2 remote sensing data. Two SO_2 peaks appear as the aircraft flies through the volcanic plume mainly consisting of SO_2 gas (bottom). SO_2 is distributed into the atmosphere and is rapidly detected by the OMI instrument on the TERRA satellite, showing the diffusion of the SO_2 from the Turrialba Volcano plume into the atmosphere (Top). The scale shown here is in Dobson Units (DU) which is the column abundance (i.e., number of gas molecules/cm^2 in the atmospheric column; 20 DU is approximately equivalent to 0.6 g/m^2 of SO_2).

The prototype ULISSES system was tested during 2009 and early 2010 in both ground and airborne application at Turrialba Volcano. This volcano was previously dormant, then that started intermittent fumarolic activity 15 years ago. During the last 2 years has undergone a series of dramatic changes with increased gaseous activity, killing most vegetation in a radius of 1 km from the crater and raising the temperature at fumaroles to more than 200 °C. A new phase of eruptive activity began on 5 January 2010 with a series of phreatic eruptions that emitted ash and gas over a large portion of Costa Rica for several weeks. The eruptions created a new large vent from which ash and high gas flux formed a visible plume going up to the boundary layer and then was carried west-southwest by the predominant winds.

Several aircraft advisories were released due to the high ash and SO_2 content of the volcanic plumes. Beginning about midnight local time on 7 January, on the basis of pilot reports and GOES-12 satellite data, the US NOAA Washington Volcanic Ash Advisory Center (VAAC) issued a Volcanic Ash Advisory notice was for Turrialba Volcano, informing aviators ash at 18,000 feet ASL, predicted to be headed to the west at about 15 kts. By 8 January, a light ash plume was observed extending about 65 km from the volcano at an altitude of about 12,000 feet ASL, moving to the west–southwest at about 10–15 kts, becoming 35 km wide at the Pacific Coast. Washington VAAC advisories continued until the morning of 9 January 2010 with additional advisories [23] concerning light gas and ash emissions on 16 January 2010.

The ULISSES test flights with a Cessna 206 Stationair aircraft departed from Juan Santamaria International Airport (SJO) in Costa Rica, about 35 km away from the volcano, flying over the summit of Turrialba Volcano in circles at different altitudes, sometimes crossing the active plume with the aircraft as depicted in Fig. 2. *In situ* VAMS ion signals for helium (He), carbon dioxide (CO_2), sulfur dioxide (SO_2), hydrogen sulfide (H_2S), plus signals for air and water were acquired during the flight as well as geo-location data with a handheld GPS unit (Garmin GPSMAP 76Cx). Flights ranged from 30 min to 2 h in duration, flying up to an altitude of 4.6 km ASL.

One of the passenger seats on the Cessna 206 aircraft was removed and the ULISSES system was placed inside the cabin to be controlled by the mission specialist. The systems inlet was connected to an external intake attached to the wing support through a 2 m long, 1/4 in. OD BEV-A-LINE tubing.

The system was calibrated at the lab prior to each deployment using a sample deliverable system with flow and pressure control and three certified NIST-traceable calibration gas cylinders provided by AIRGAS/PRAXAIR in Costa Rica for zero-test-span calibration points.

Several flights were performed months before (April, September, and October 2009) and days after the 5 January 2010 eruption. The volcano was still very active during the last deployment on 19 January 2010 described in this paper. Remote sensing data acquisitions using satellite platforms were requested and performed with the ASTER (Advanced Spaceborne Thermal Emission and Reflection Radiometer) carried by the NASA TERRA satellite, and the Ozone Monitoring Instrument (OMI) on board the NASA AURA satellite. OMI-SO_2 retrievables were downloaded for comparison with *in situ* MS data.

In conjunction with the airborne campaigns, a series of ground visits to the crater were conducted on the same day the airborne data were collected, to allow comparisons between ground and airborne *in situ* measurements.

ULISSES gas data, GPS geo-location and temperature data were collected during site visits to several fumaroles on the main active crater. The 19 January visit was performed under Costa Rican National Emergency Commission (CNE) authorization allowing no more that 1 h of exposure time and minimal research personnel at the site due to the high risk of a new phreatic eruption. The ULISSES system was transported as a backpack for easy operation (Fig. 3) and placed near the selected fumaroles. Operators with full face masks and protection gear walked to the site to perform the *in situ* measurements. Ground-based gas measurements were done in the same location before and after the eruption, coincidentally being only 30 m away from a new vent which opened with the 5 January eruption. Gas flow was extremely high at this new vent, (with sound levels comparable to that of a jet engine), and temperatures were greater than 500 °C, so it was not possible to measure gas coming out from this new vent. Observations, however, were carried out on nearby fumaroles with temperature reaching 90–100 °C.

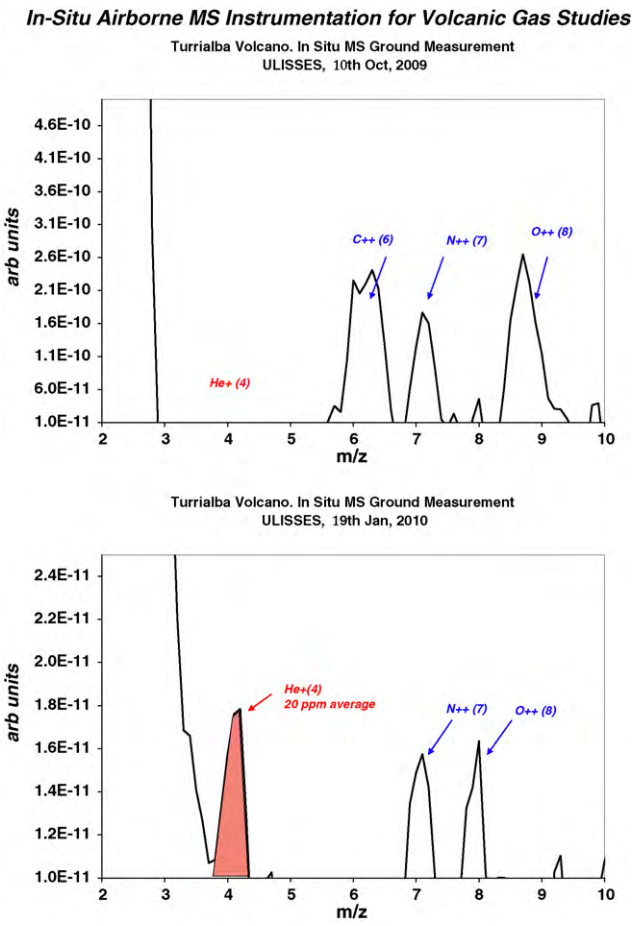

Fig. 7. ULISSES ground data from Turrialba Volcano taken at fumaroles near main vent before (top) and after (bottom) the 5th January 2010 eruption. Mass spectra shows a 20 ppm average helium emission after the eruption, not seen at the previous site survey done in October 2009. Higher C++ ion signal is due to higher CO_2 presence on fumaroles before the eruption.

3. Results and discussion

The ULISSES system detected the primary gas species emitted from Turrialba Volcano at a number of fumaroles and while flying through the volcanic plume. Fig. 5 shows a 3D rendering of the flight path performed over Turrialba Volcano on September 2009, months before the eruption. Similar flight paths were conducted in April 2009 and January 2010. The last fly-over was conducted while the volcano was erupting (Fig. 4). Flying conditions were good, but small ash eruptions made the flying conditions extremely hazardous for a manned aircraft.

Fig. 5 shows the mass spectra of the volcanic plume for the January 2010 fly-over after the eruption. The measured species include water vapor (H_2O) in large quantities, carbon dioxide (CO_2), sulfur dioxide (SO_2) and hydrogen sulfide (H_2S). The mass spectra also showed large quantities of nitrogen (N_2), Oxygen (O_2) and Argon (Ar), which are common since the volcanic emissions diffuse and mix with the surrounding air envelop. The SO_2 peak at m/z 64 amu is very distinctive (around 2% concentration), even though it has already diffuse with the surrounding air. CO_2 (m/z = 44 amu) and H_2S (m/z = 34 amu with oxygen isotope subtraction) were also present in the volcanic plume. In contrast, in the pre-eruption *in situ* airborne sampling conducted in April and September 2009, the ULISSES mass spectra showed almost no sulfur dioxide nor hydrogen sulfide content, and only water vapor and carbon dioxide as primary gas species in the plume.

To validate the sulfur dioxide findings, an image generated from the OMI instrument onboard the AURA satellite showing the SO_2 column content in the atmosphere for the same day is shown in Fig. 6. The remote sensing image shows a very large atmospheric SO_2 signature, correlating well with the high SO_2 content obtained with the *in situ* airborne mass spectrometer and originating from the Turrialba Volcano eruptive activity.

In situ MS flights were conducted in early hours of the morning when cloud coverage and flying conditions are best for plume sampling. After landing, the ULISSES instrument was taken to Turrialba crater for ground *in situ* fumarole analysis. The fumarole sampling showed similar qualitative results as compared to the airborne measurements, but VAMS components were in much higher concentration than the concentration levels obtained in the air due to the low air mixing ratios occurring at the fumaroles' site. For the pre-eruptive sampling done in April and September 2009, besides water vapor, carbon dioxide was the dominant gas. SO_2 was present, but in concentrations 10 times lower than CO_2. For samples taken during eruption conditions in January 2010, this ratio reverted and SO_2 contents in some cases reached almost 50% concentration in fumarolic emissions, while CO_2 remained in the low single digits percentage. In the case of helium, this VAMS gas was not present in the measurements performed before the eruption,

but a detectable quantity of 20 ppm on average, as shown in Fig. 7, was detected in data from the January 2010 ground deployment while the volcano sustained eruption activity.

These data are encouraging in the context of detecting gas signal precursors to volcanic eruptions. Sometime between the September 2009 and January 2010, the helium signal changed from non-detectable to 20 ppm on average. Our working hypothesis is that helium was degassed from fresh magma entering the magmatic supply system, contributing to pre-eruption gas pressure buildup. Driven by gas buildup and overpressure within the system, a failure threshold was exceeded, and the gas-driven eruption began. Clearly, during the initial overpressure phase, it would be possible for highly volatile helium to seep toward the ground surface, and that precursory release is what we believe we detected in the case of Volcan Turrialba. More generally, continuous *in situ* MS measurements could potentially document such important and related precursory changes in gas geochemistry not only for helium, but for a variety of species, and would be useful as part of an eruption warning system.

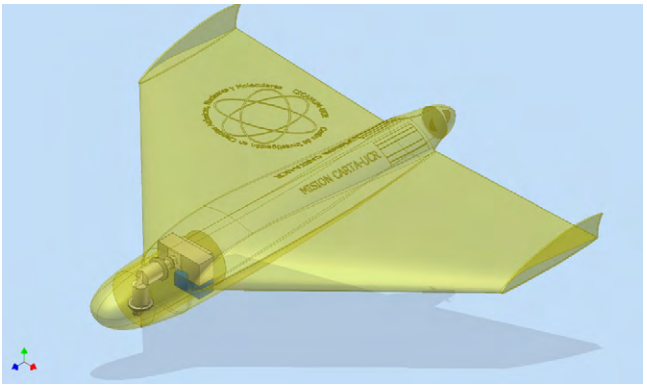

Fig. 8. Preliminary isometric diagram of UAV-based *in situ* mass spectrometer system for the study of Costa Rican volcanoes using an electric UAV platform (Carolo D-150 from Mavionics) plus miniature mass spectrometer and miniature turbopump system.

4. Conclusions and future development

In summary, the ULISSES instrument is an *in situ* harsh-environment MS-based system of about 10 kg, a volume around 21,000 cm^3 and capable of monitoring multi-gas species simultaneously, within a mass range from 1 to 100 amu. It is targeted to measure volcanic plumes both at ground level and from airborne platforms to better assess and understand VAMS signals as precursors to eruptive activity. The current detection limits of the instrument are below 10 ppm for permanent gases with a typical accuracy of 10%. The system requires an operator, is rugged, and can operate up to an altitude of 3.8 km ASL for up to 4 h.

The field deployments to Turrialba Volcano, both ground and airborne, before and after the 5 January 2010 eruption demonstrated the usefulness of the ULISSES instrument to characterize the volcanic gaseous emission concentrations in harsh-environment conditions. The two key achievements of the present study were the confirmation of helium presence after the eruption, as well as the airborne MS measurements of SO_2 allowing the possibility of correlating *in situ* data with satellite remote sensing measurements from OMI and ASTER [18].

The deployment also showed areas of improvement for the MS system: better resolution at low masses so zero blast and hydrogen signal does not affect the helium signal, higher sensitivity for SO_2 to be able to detect in the ppb levels. An automatic inlet pressure regulation and autonomous calibration subsystems are on the list of components to incorporate, taking weight and size into account as well.

The data provided by this system is a key step towards a better understanding of geophysical phenomena surrounding eruptive activity. The combination of these gas measurements and imaging data allows the quantitative mapping of gas concentration at volcanic structures and serves as a tool to assess hazardous conditions by visualizing and modeling possible scenarios of volcanic activity. In addition, such gas sampling flights and subsequent correlation with remote sensing data, provide otherwise unobtainable, sub-orbital, *in situ* calibration and validation data for SO_2 retrievals from existing NASA orbital instruments, such as ASTER, OMI, MODIS (Moderate Resolution Imaging Spectroradiometer), and AIRS (Atmospheric Infrared Sounder). Such measurements and observations are highly relevant to aviation safety and global climate assessments. These data are also highly pertinent to the development and implementation of the NASA HyspIRI hyperspectral visible and multispectral thermal infrared orbital instrument, now under study and development as one of the next generation Earth Science missions as recommended by the U.S. National Research Council (NRC) Decadal Survey on Earth Science and Applications from Space.

These ULISSES airborne measurements also yielded insights relevant to the use of UAVs as future airborne measurement platforms (Fig. 8), in order to improve overall deployment cost, to lower risks, and to increase *in situ* spatial resolution. The target is to create an airborne UAV-MS system capable of rapidly and cost effectively collecting high caliber, low altitude, *in situ*, multi-gas characterization data near high-risk volcanoes, under flight conditions deemed too hazardous to manned aircraft—essentially to collect data that are otherwise impossible to obtain at this point. Using UAVs for *in situ* sampling removes aircrew risks and allows the use of lower cost platforms versus manned aircraft [19,20]. Currently, very few sub-orbital (airborne) data on volcanogenic gas concentration exist for validating and calibrating instrumental data and phenomenological models for current and future gas sensing space missions. The UAV-MS package would provide a robust *in situ* sampling capability and data from hazardous flight environments crucial to the calibration and validation of instruments and volcanologic models, and fills an *in situ* data gap for volcanic plumes and clouds.

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