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# Toward Rapid balloon Experiments for sudden Aerosol injection in the Stratosphere (REAS) by volcanic eruptions and wildfires

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**ABSTRACT:** Stratospheric aerosols are greatly influenced by medium-to-large volcanic eruptions. Over the last few years, extreme wildfires have been identified as new sources of stratospheric particles, in the form of carbonaceous aerosols injected by pyroCb events in the upper troposphere and lower stratosphere, associated with significant impacts on climate and ozone chemistry. To assess the impact of wildfires and volcanic eruptions on stratospheric aerosol loadings in the Northern Hemisphere, the Rapid Experiments for sudden Aerosol injection in the Stratosphere project has been initiated. REAS is an international initiative that aims to respond to sudden events impacting stratospheric aerosol composition. Seventeen balloons were launched from Reims, Eastern France, between November 2021 and January 2022 to quantify the atmospheric content for both aerosols and trace/greenhouse gases from the ground up to stratospheric levels. The main measurements concerned trace gases ( $\text{CO}/\text{CO}_2$  as tracers of smoke) and aerosol together with ozone using instruments such as a gas collector, optical particle counters, backscatter sondes, an aerosol sampler, an aerosol impactor, and ozonesondes. GSMA launch facility provided unique possibilities of combining multiple measurements in one flight thanks to medium flights (corresponding to a 6kg payload). While no major event impacted the stratosphere during the campaign, we particularly discuss the influence of the aged volcanic plume from La Soufrière volcano (Saint Vincent island) and smoke particles from series of pyroCb events that took place in North America. The burden as well as the optical and microphysical properties of the observed aerosols are quantified from these in situ observations in association with various satellite data.

## 1. Introduction and Science Motivation

The stratospheric aerosol layer is a key component of the earth climate system through its interaction with radiation, ozone chemistry and its influence on cirrus cloud formation in the upper troposphere (Kremser et al. 2016). Major volcanic eruptions can affect stratospheric aerosol loadings for years with subsequent surface temperature cooling as shown after Mt Pinatubo eruption in 1991 (McCormick et al. 1995). However, smaller but more frequent volcanic eruptions can also significantly influence the optical, physical and chemical properties of the stratospheric aerosol layer (Vernier et al. 2011). The stratospheric aerosol layer has been studied for decades with optical particle counters launched onboard large balloons by the University of Wyoming (Deshler et al. 2006) and through LiDAR (Light Detection And Ranging) observations from multiple locations (Jäger 2005). In addition, satellite observations from the Stratospheric Aerosol and Gas Experiment (SAGE) missions provide one of the longest records of stratospheric aerosol optical properties since the mid-70s and were used to develop long-term databases such as the Global Space-based Stratospheric Aerosol Climatology (Kovilakam et al. 2020). Figure 1 shows how the stratospheric aerosol layer in the Northern Hemisphere has been affected by major and moderate volcanic events since the late 70's.

Over the last decade, major wildfires affected the stratosphere at levels never observed before. The 2017 British Columbia wildfires and 2019/20 Australian bushfires affected the stratosphere for several months resulting in Stratospheric Aerosol Optical Depth comparable to medium volcanic eruptions (Torres et al. 2020; Lee et al. 2023; Peterson et al. 2021; Khaykin et al. 2020). Due to the difference in their optical properties, volcanic and wildfires plumes have different impacts on chemistry and climate. The absorbing nature of smoke produces significant heating in the plume with induced self-lofting mechanisms augmenting aerosol lifetime and prolonging the impacts. Due to the increased complexity of the stratospheric aerosol layer impacted by multiple sources, better characterizing its optical, physical and chemical properties is fundamental. While aircraft observations can provide detailed information with a wealth of measurements, limitations including cost, deployment readiness and cruise altitudes make it challenging to respond to unpredictable events. Rapid balloon deployments of lightweight sensors remain an affordable and manageable way to provide critical information about plume properties. Several volcano response activities have been initiated through the Stratospheric Sulfur and Its Role in Climate (SSiRC) (Vernier et al.

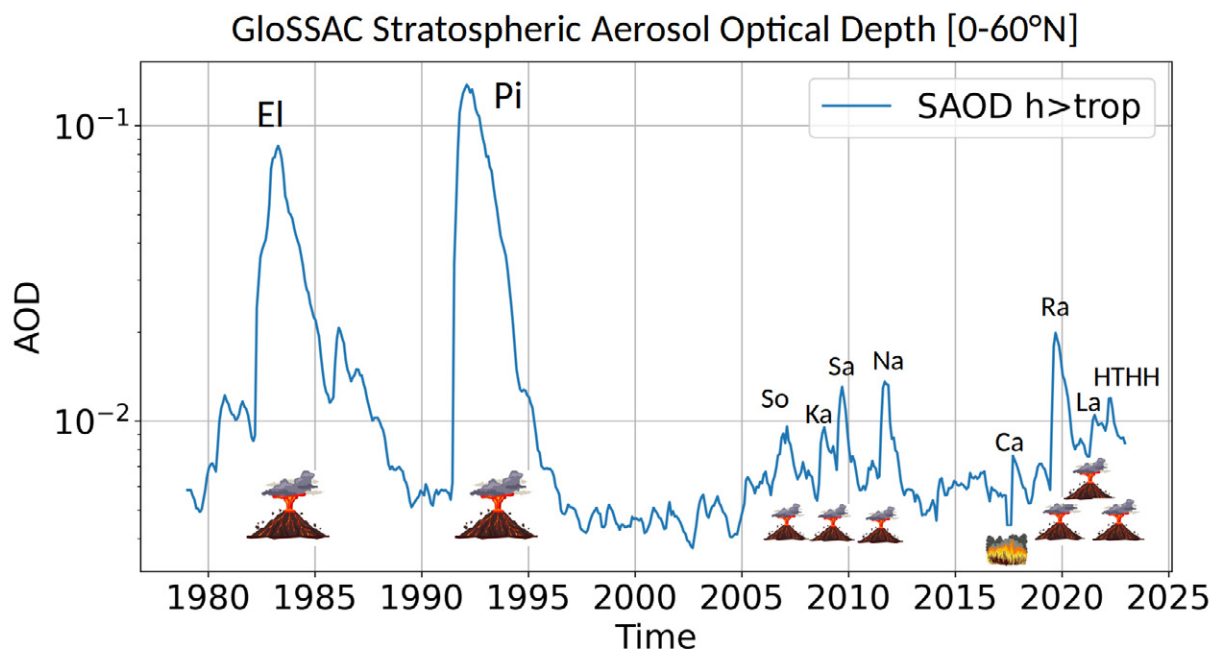


FIG. 1. Stratospheric AOD Evolution and influence of major and moderate volcanic events since 1975 for the Northern Hemisphere. The El Chichon, Pinatubo, Soufrière Hills, Kasatochi, Sarychev, Nabro, Raikoke, La Soufrière and Hunga Tonga-Hunga Ha-apai volcanic eruptions are labelled as 'El', 'Pi', 'So', 'Ka', 'Sa', 'Na', 'Ra', 'La' and 'HTHH' respectively. 'Ca' refers to the 2017 Canadian wildfire event.

2023) and by NASA (Carn et al. 2022) already rely on the rapid deployments of balloon-borne sensors. The REAS initiative aims to deploy and test new instruments to make rapid and long-term measurements in volcanic and wildfire plumes in the Northern Hemisphere. The first test campaign took place during the fall/winter 2021 and will be described in this paper.

## 2. Atmospheric conditions before the first REAS campaign

Here we describe significant events that have impacted aerosols and greenhouse gases atmospheric content in Northern Hemisphere during 2021.

### a. La Soufriere eruption

La Soufrière volcano (13.33N; 61.18W) located on the island of St Vincent in the Caribbean erupted on 9 April 2021 with multiple explosive events until 22 April 2021. Using the Advanced Baseline Imager on the Geostationary Operational Environmental Satellite (GOES), and the In-

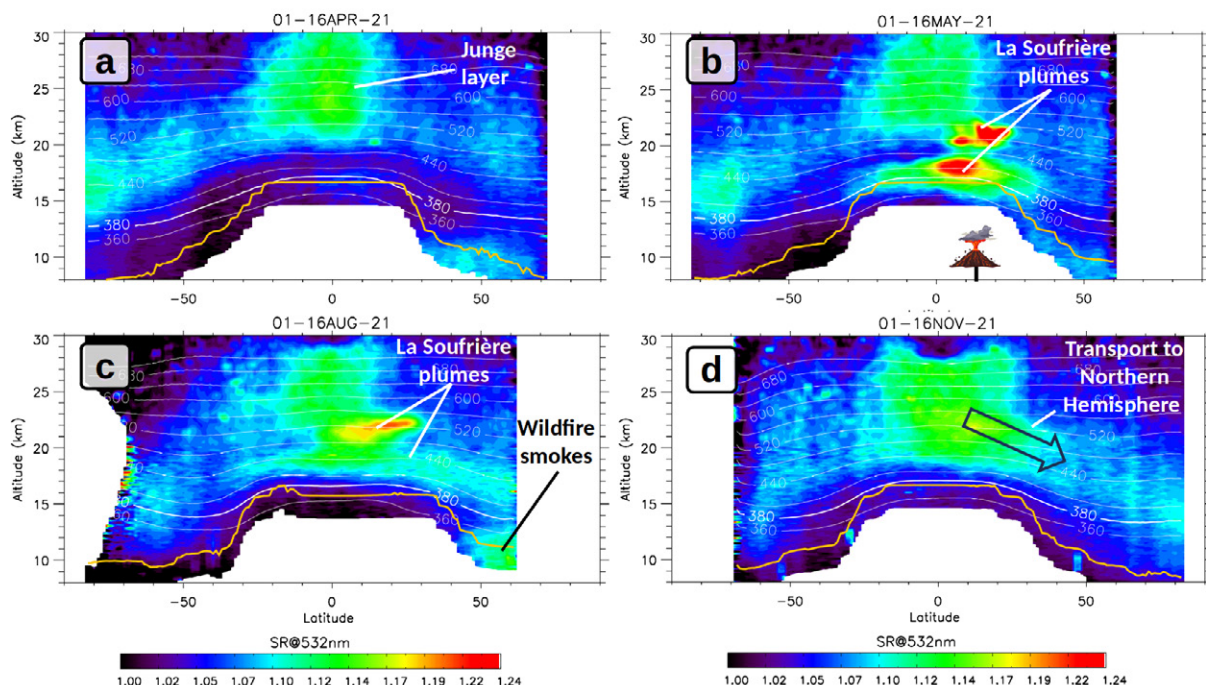


FIG. 2. Zonal mean cross-section of Scattering Ratio (SR) from CALIOP level 1 V4.01 between 01-16 April 2021 (a), 01-16 May 2021 (b), 01-16 October 2021(c) and 01-16 November 2021 (d). White lines are zonal mean isentropic surfaces, and the tropopause altitude is shown in orange. Data 3 km below the tropopause are discarded for possible contamination by cirrus cloud)

frared Atmospheric Sounding Interferometer (IASI), Taylor et al. (2022) showed that the eruption went through 32 explosive sequences with at least 32 injections in the upper troposphere and lower stratosphere between 13 and 19 km associated with a total  $\text{SO}_2$  emitted of  $0.57 \pm 0.44$  Tg. The plume reached 40 degree N by the end of April. Figure 2 shows the zonal monthly mean SR cross-section derived from CALIOP level 1 V4.01 product (Vernier et al. 2009) after screening clouds in the UT using a depolarization ratio of 5% and removing the data 3 km below the tropopause. The white potential temperature isentropic surfaces are also plotted together with the tropopause level in orange. The multiple volcanic layers produced after La Soufrière eruption are visible between 1-16 May and between 14 and 21 km. Consistent with Taylor et al. (2022), the plume is dispatched across the upper troposphere and lower stratosphere due to the multiple injections reported during the eruption. The lower part of the plume is transported between 380 and 420 K at mid-latitudes between May and August 2021. The cross-section between 01-16 November indicates a continuation of this transport, possibly up to 520 K.

### *b. Multiple wildfires across the Northern Hemisphere in spring and summer 2021*

In 2021, major wildfires sparked across North America and Siberia and plumes may have reached Europe on several occasions. Prior to the REAS campaign, smokes from California wildfires which took place from August to September 2021 could be observed on September 13<sup>rd</sup> in France, during a high altitude balloon flight launched from the GSMA facility (see Sidebar). Figure 3a shows the vertical concentration profiles of CO<sub>2</sub>, CH<sub>4</sub> and CO obtained with an Aircore atmospheric sampler on that day (Karion et al. 2010; Membrive et al. 2017). A CO profile from the Copernicus Atmospheric Monitoring Service (CAMS) monthly mean analysis of November 2016 is also plotted as a reference since Europe was not impacted by wildfire events in 2016 (see Section 5). Vertical profiles show an increase in CO and CH<sub>4</sub> concentrations between 6.5 and 11 km, as well as two distinct levels of CO<sub>2</sub> concentrations at the same altitudes (shown in cyan and magenta on Figure 3). This difference in concentration suggests the presence of air masses of different origin. Back-trajectories from the two median levels (7.5km and 10km) were calculated using HYSPLIT trajectory software (Stein et al. 2015) and show that one of the air masses comes from a low-lying area different from the first (Figure 3b). Using the Ozone Mapping and Profiler Suite-National Polar orbiting (OMPS)/Partnership (NPP) Aerosol Index swath orbital V2 (Torres 2019) associated with air mass back-trajectories, it appears that the two trajectories encountered different plumes on September 4th and on September 5th as shown in Figure 3c and 3d (thicker lines correspond to the position of air masses on a given day). On September 9<sup>th</sup>, low altitude air masses may have been uplifted from about 4km to 10km thanks to synoptic lift above Canada.

Storms induced by wildfires also known as pyroCbs have been observed for several decades (Fromm et al. 2022). Through the pyroCb Information Exchange (<https://groups.io/g/pyrocb>), between June 2021 and October 2021, at least, four of them were identified with subsequent stratospheric impacts (see Supplemental material for detailed description). Among them, the KNP Complex fire, which was the result of two merged fires in the Sequoia & Kings Canyon National Park, produced a series of pyroCbs on October, 4<sup>th</sup> and it might have impacted both aerosol and GES over Europe as shown on Figure 4. Figure 4a presents an RGB composite image from Sentinel 3A OLCI taken on October 4<sup>th</sup> where one of the pyroCbs can be identified. On October 10<sup>th</sup>, a plume at about 13km high could be seen on CALIPSO data (Figure 4b) and back-trajectories from

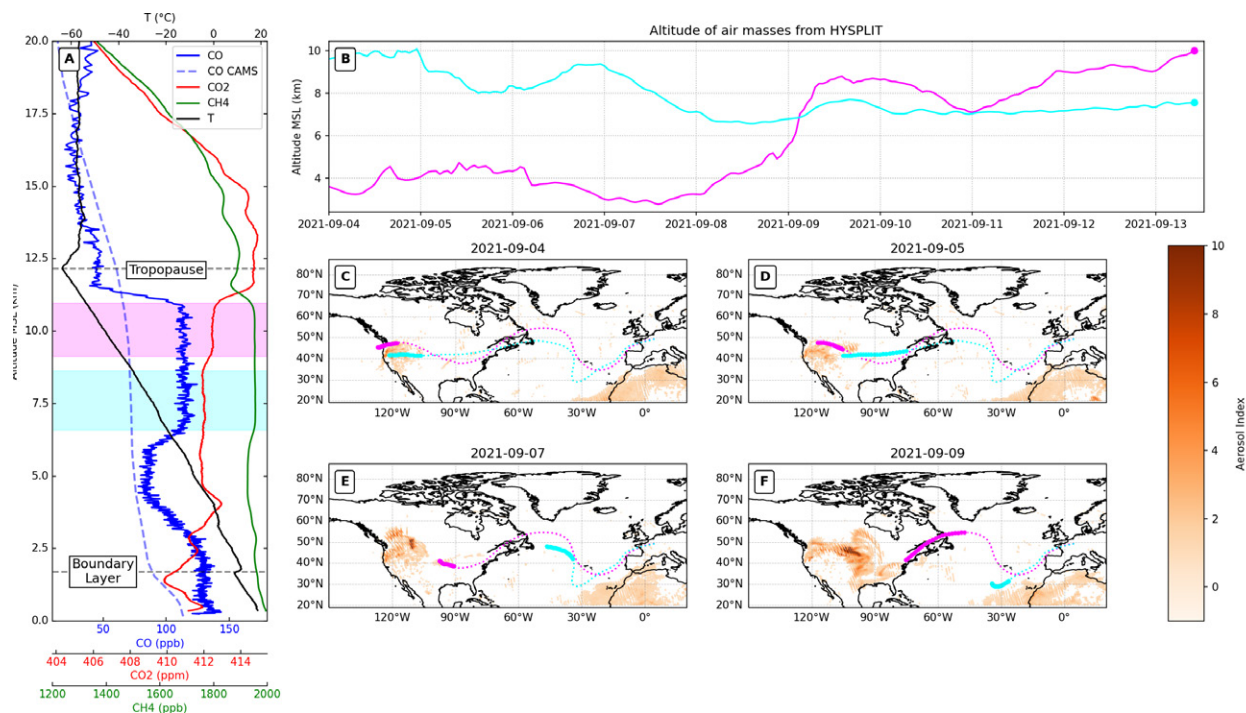


FIG. 3. Influence of US wildfires on tropospheric trace gas concentrations in Europe (prior to the REAS campaign). (a) Vertical profile of CO, CO<sub>2</sub> and CH<sub>4</sub> associated with temperature with two probable air masses (in cyan and magenta) on September 13<sup>rd</sup>. (b) Vertical evolution of the identified air masses calculated from HYSPLIT. (c-f) OMPS-SNPP Aerosol index and possible air mass position (thick lines) from HYSPLIT from September, 6<sup>th</sup> to September, 9<sup>th</sup>.

HYSPLIT indicate that the smoke could have transported from the KNP Complex fire to Europe (Figure 4c).

In order to study the influence of a moderate volcanic eruption and multiple wildfires in the Northern Hemisphere, the REAS project was created by joining the expertise of several laboratories in Europe and in the US to combine aerosol and trace gas measurements. It represents a unique opportunity to assess the various sources, the burden, and the physical/chemical properties of stratospheric aerosols driven by volcanic eruptions and wildfires. In this paper, we will describe the logistics, the infrastructure, the payloads and the preliminary results of the REAS project.

### 3. Campaign preparation and planning

Scientific ballooning requires extensive preparation and evaluation of risks to the public, the military and aviation even when small payloads (~5 kg) are involved. Here we summarize the logistical challenges for those flights.

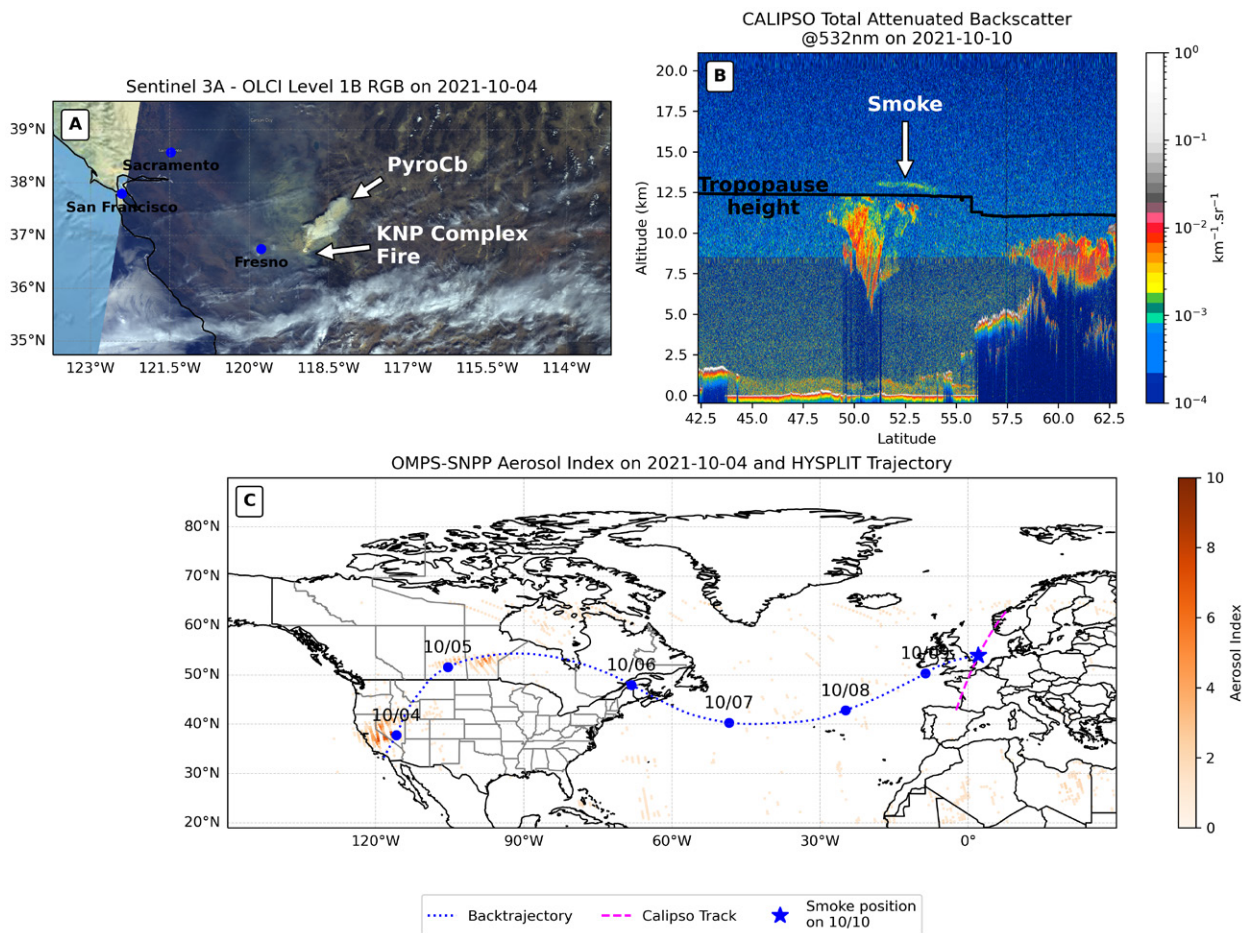


FIG. 4. (a) RGB composite image of pyroCb on October 4<sup>th</sup> (b) CALIPSO Total Attenuated Backscatter at 532nm showing a plume in Europe, (c) HYSPLIT back-trajectory associated with OMPS Aerosol index measured on October 4<sup>th</sup>

#### a. Flight Management Tools and Procedures

Weather balloons cannot be controlled horizontally as their trajectories are completely subjected to winds. However, getting accurate predictions of the scientific payload landing area is important, even critical when flying over populated areas. To ensure maximum safety for both people and instruments, a set of procedures has been developed, together with instrumental tools and software (see section below). During the REAS campaign, medium balloon flights were also conducted following a new procedure defined in collaboration with air traffic control.

(i) *Flight procedure and trajectory simulations* Several models for trajectories previsions have been developed over the years (Conner and Arena 2010; Sóbester et al. 2014; Lee and Yee 2017; Robyr et al. 2020). Among them, the CUFS Predictor from Cambridge University is very popular

as it provides a web interface for trajectory predictions (Snowman et al. 2013). We slightly adapted the model to use winds from ECMWF's IFS forecasts (Owens and Tim 2018) instead of the original inputs from NOAA's GFS forecasts. The former is more accurate over the European region (Martineau et al. 2016; Hoffmann et al. 2019) and simulations compared to flight trajectories showed differences less than 5 km between effective and predicted landing points.

Thus, before any flight, trajectory simulations are performed to determine the best suited ascent rate and flight altitude interruption. To approve the launch, simulations were performed 48h and 24h before the flight, and on the day of the flight.

Since the balloon launch and landing may occur in populated areas and military sites, we have defined two criteria to proceed for a balloon launch: round wind must be less than  $5\text{m.s}^{-1}$  and predicted landing area must not be within 10 km of a small town (population  $\leq 10000$ ), military site or airport.

(ii) *Communicating separator and I.R.M.A* The use of a communicating separator is a key feature of the flight: it regularly transmits the payload's position and altitude using satellite communication (every 2 or 4 minutes) and terminates the flight by cutting the suspension rope between the balloon and the flight train using a hot wire. The ascent termination can be programmed at a given altitude, after a scheduled delay, or remotely activated. The separator is also associated with a web interface called I.R.M.A (Iridium Remote Monitoring Application) which allows real time visualization of payload position and predicted landing area (Figure 5a). Using 4G tablets, recovery teams can access the I.R.M.A website to pre-position themselves as close as possible to the landing zone and quickly retrieve the scientific payloads (Figure 5b).

### *b. Medium Flights*

In Europe, unmanned free balloon flights must comply with Annex 2 of the "Standardised European Rules of the Air" (Council of European Union 2021). This annex defines three different types of balloons (light, medium and heavy balloons) for which different rules apply. Most scientific flights belong to the light balloons category as regulations are simplified and allows one or more packages to be carried with a combined weight of less than 4 kg. Medium balloons are subject to more constraints than light balloons but the combined weight can reach 6 kg. In both cases, one single package cannot weigh more than 3 kg. The main advantage of medium balloons

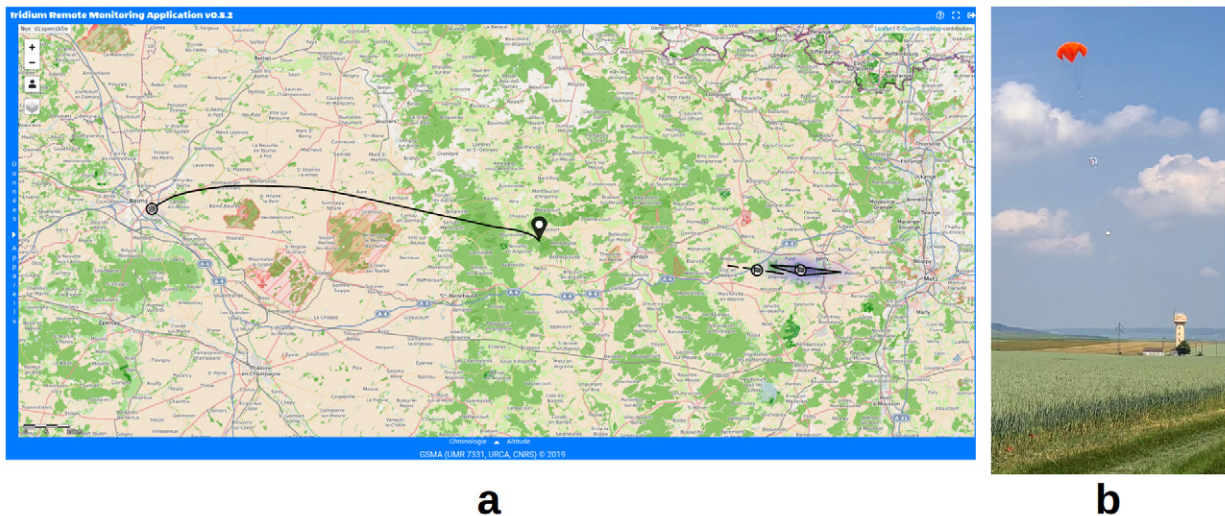


FIG. 5. (a) Web interface of I.R.M.A with current flight trajectory in black: dashed area represents landing area if the flight is terminated immediately and continuous line area represents nominal landing area for a 29km altitude flight. (b) Example of a descending flight train image taken by the recovery team while already close to the landing site using I.R.M.A trajectory forecasts.

is that they can allow instrument inter-comparison or multiple profiles acquisition in one single flight. However, they are considered by air control as potentially dangerous for airline traffic; therefore, flight rules are more restrictive. We have been actively working with the air safety authorities to develop a flight protocol for launching this class of balloon. This protocol includes a 24h notice before any launch with detailed trajectory forecasts including visualization of areas where the balloon flights under specific flight level (FL-315 corresponding to 31500ft) as well as 2D representation of trajectories on the International Civil Aviation Organization (ICAO) flight information region maps. A NOTAM (notice to airmen) is also issued from the air traffic authorities over the period of the campaign.

#### 4. The REAS Campaign

The REAS Campaign took place from the GSMA facility in Reims, between November 2021 and January 2022. As the first campaign of this project, NIA, NASA Langley, LPC2E and GSMA teams selected readily available payloads that measure greenhouse gases and aerosol optical and micro-physical properties. We also flew new sensors including an aerosol impactor and an aerosol sampler equipped with a variety of filters (PTFE or carbon-based) for targeting specific aerosols

and organic compounds. They are listed in Table 1 and a detailed description is available in Supplemental material.

and

REAS Payloads	Measured parameters	Measurement technique
M10	Pressure, Temperature, Humidity, Latitude, Longitude Altitude, Wind speed and direction	Piezoresistive, thermistor, capacitive
M20	Pressure, Temperature, Humidity, Latitude, Longitude Altitude, Wind speed and direction	Piezoresistive, thermistor, capacitive
iMet	Pressure, Temperature, Humidity, Latitude, Longitude Altitude, Wind speed and direction	Piezoresistive, thermistor, capacitive
O3 ECC	Ozone concentration	Electrochemical
Aircore	CO <sub>2</sub> , CH <sub>4</sub> and CO concentration	Air sampling
SAMPLER	Chemical composition of aerosol	Aerosol filtering
IMPACTOR	Chemical composition of aerosol	Aerodynamical impaction
LOAC	Aerosol concentration and distribution (19 size bins, 0.2-50 $\mu$ m)	Laser counting
POPS	Aerosol concentration and distribution (16 size bins, 0.14-3 $\mu$ m)	Laser counting
POPC	Aerosol concentration and distribution (6 size bins, 0.3-10 $\mu$ m)	Laser counting
NPOPC	Aerosol concentration and distribution (30 size bins, 0.3-10 $\mu$ m)	Laser counting
COBALD	Scattering ratio at 470 and 940nm	Light scattering with diode

TABLE 1. List of the payloads used during the REAS campaign.

Seventeen balloons were launched (Figure 6) and four of them were medium balloons carrying multiple instruments (Table 2).

## 5. Campaign Results

The main goals of the REAS campaign were to: 1) characterize stratospheric aerosol properties using multiple balloon-borne instruments, 2) test in-flight new instruments like the IMPACTOR that cannot normally fly in Europe due to their weight and 3) conduct instrument inter-comparison thanks to the medium flight possibilities.

The deployment of medium flights was used to gather data from multiple instruments. This allowed us to characterize the chemical composition of aerosols from the surface to the balloon burst altitude, revealing the presence of organic material in the upper troposphere and lower stratosphere (UTLS), alongside with the aerosol microphysical properties among different vertical layers (Benoit et al. 2023).

The balloon measurements conducted during the REAS campaign were predominantly affected by the long-range transport of the La Soufrière eruption. Figure 7 shows the daily aerosol extinction (version 2 product) (Taha 2020) from the NASA/NOAA OMPS-Limb Profiler (LP) instrument over

Date UTC	Time UTC	Flight No.	Flight class	Maximum altitude	Payloads	Payload weight	Balloon type
2021/11/18	10h09	REAS01	Light	8,2km	POPS, LOAC, M10	2,5kg	1200g
2021/11/24	10h07	REAS02	Light	29km	Aircore, M20	3,1kg	1600g
2021/11/24	19h00	REAS03	Light	27,5km	POPC, COBALD, iMet	2,6kg	1200g
2021/11/25	09h50	REAS04	Light	8,7km	POPS, LOAC,M10	2,5kg	1200g
2021/12/02	19h00	REAS05	Light	29km	POPC, COBALD, iMet	2,6kg	1200g
2021/12/08	13h20	REAS06	Light	16km	POPC, iMet	2,5kg	1200g
2021/12/08	13h25	REAS07	Medium	16,1km	IMPACTOR, iMet	5,6kg	1600g
2021/12/16	9h24	REAS08	Medium	27,5km	LOAC, POPS, AirCore, M10	5,3kg	1600g
2021/12/16	9h30	REAS09	Medium	27,1km	IMPACTOR, iMet	5.6 kg	1600 g
2021/12/16	9h27	REAS10	Light	27km	SAMPLER, POPC, iMet	3.8 kg	1600 g
2021/12/22	9h28	REAS11	Light	17,5km	POPC, M20	2.2kg	1600g
2022/01/06	9h38	REAS12	Light	23,4km	POPC, NPOPC, O3, iMet, M10	3,1kg	1200g
2022/01/06	9h41	REAS13	Light	25km	SAMPLER,iMet	2.8kg	1200g
2022/01/17	19h00	REAS14	Light	26,3km	POPC, NPOPC, COBALD, iMet	2,8kg	1200g
2022/01/18	09h11	REAS15	Light	25,5km	Aircore, Ozone ECC, M10	3.9kg	1600g
2022/01/18	09h08	REAS16	Medium	25,6km	POPC, NPOPC, SAMPLER, iMet	5,2kg	1600g
2022/01/18	14h00	REAS17	Light	20km	SAMPLER, iMet	2.8kg	1200g

TABLE 2. List of balloon flights during the REAS campaign together with the date, time, flight type, payload, weight, and balloon size information. Note that REAS02 flight was also a part of monthly Aircore launches done for the MAGIC initiative (Crevoisier et al. 2019).

Europe. The top image gives a general view of the stratospheric aerosol content from 2016 to end of 2022. Year 2016 could be used as a background level as no major event impacted Europe. The stratospheric aerosol content over the 2021-2022 period is characterized by different plumes i.e., from the Raikoke volcano with a signature remaining in early 2020, from fires possibly from North America and/or Siberia in fall 2021 and spring/summer 2022, and from the Hunga Tonga volcano in late 2022.

Figure 7 (bottom) depicts a noticeable increase in the aerosol content below an altitude of 20 km during the mid-2021 to early 2022 timeframe, in contrast to the conditions observed in early 2021, as observed by the SAGE III/ISS space-borne instrument (NASA/LARC/SD/ASDC) and in agreement with OMPS-LP data. The higher vertical resolution from SAGE III depicts a double peak structure (e.g., in January 2022 near 12.5 and 17.5 km) likely pointing to different injection processes and/or air mass origins in the lowermost stratosphere possibly due to long-range transport of fire smoke.

These results are consistent with in situ balloon-borne observations conducted during the REAS campaign. Figure 8 shows concentration profiles for sizes greater than  $0.15\mu\text{m}$  obtained by the

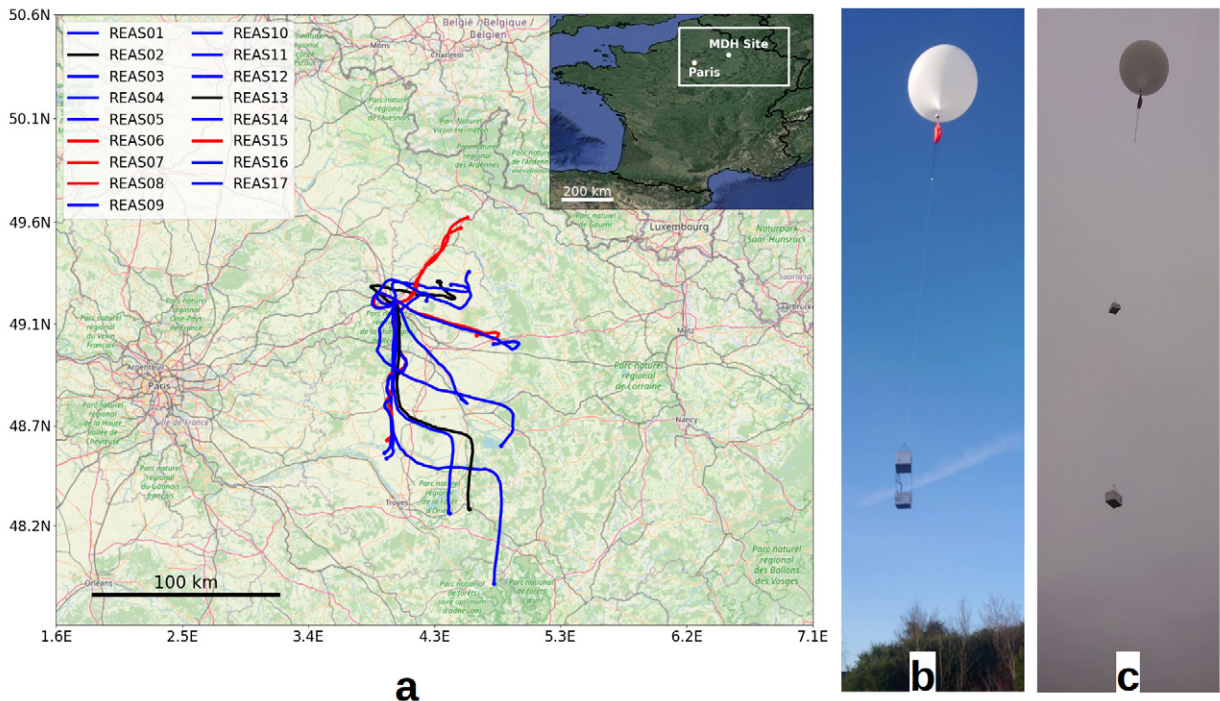


FIG. 6. (a) Geographic localization of MDH Site with all flight trajectories: **blue lines** correspond to light balloons, **red lines** to medium balloons and **black lines** to night flights, (b) Medium balloon with Impactor (REAS06) and (c) Medium balloon with POPC, NPOPC and Sampler (REAS15)

POPC and POPS optical particle counters and three Scattering Ratio (SR) profiles observed by COBALD (Brabec et al. 2012; Vernier et al. 2016) during nighttime in collocation with POPC. Profiles observed in the stratosphere by the POPC and POPS instruments are consistent in terms of concentration values. The boundary layer is visible on both instruments below 1.5 km followed by a minimum of aerosols in the free troposphere and an increase in the stratosphere peaking between 12.5 and 20 km and followed by a gradual decrease in the mid-stratosphere. The profiles obtained simultaneously from the POPC and COBALD instruments are also consistent with each other with similar profile shapes. The small structures are also well reproduced by both instruments such as a peak in aerosol concentrations and SR near 14 km and a minimum near 12 km on 01/17/22. Between 10 and 13 km, both POPC and COBALD show a decrease in aerosol concentration/SR from 11/12/21 to 12/02/21 and on 01/17/22. The vertical structure of the aerosol clearly fluctuates over the period of the REAS campaign possibly reflecting a still non-homogenized aerosol content with the presence transient layers and/or the effect of microphysical processes like sedimentation

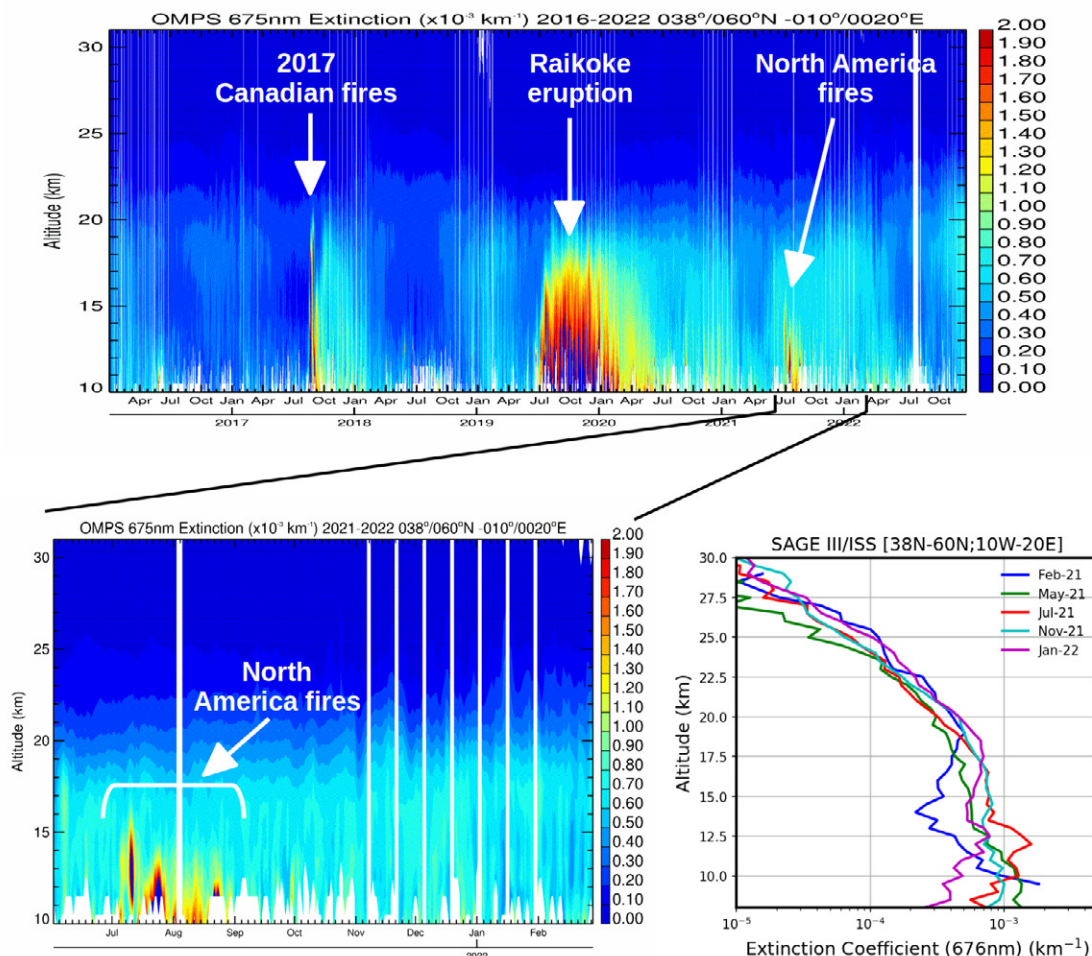


FIG. 7. OMPS-LP time series of daily mean aerosol extinction at 675nm between January 2020 and November 2022 above the 38°N-60°N/10°W-20°E area corresponding to the major part of Europe (Top). Zoom of OMPS-LP extinction time series from June 2021 to February 2022 (Bottom left) for the same area. Vertical profiles of aerosol extinction from the SAGEIII/ISS instrument (Bottom right).

(Benduhn and Lawrence 2013; Sukhodolov et al. 2018) which will have to be investigated with chemistry-transport model simulations

The growing influence of extreme wildfires on the stratosphere observed over the past few years marks a profound shift in our understanding of the impacts of climate changes on the stratosphere (Fromm et al. (2022) and references therein). The combination of trace gas measurements such as CO, CO<sub>2</sub>, CH<sub>4</sub>, and ozone with aerosol optical and microphysical properties are helpful to identify smoke plume layers at stratospheric levels. Figure 9 is an example of Aircore and aerosol

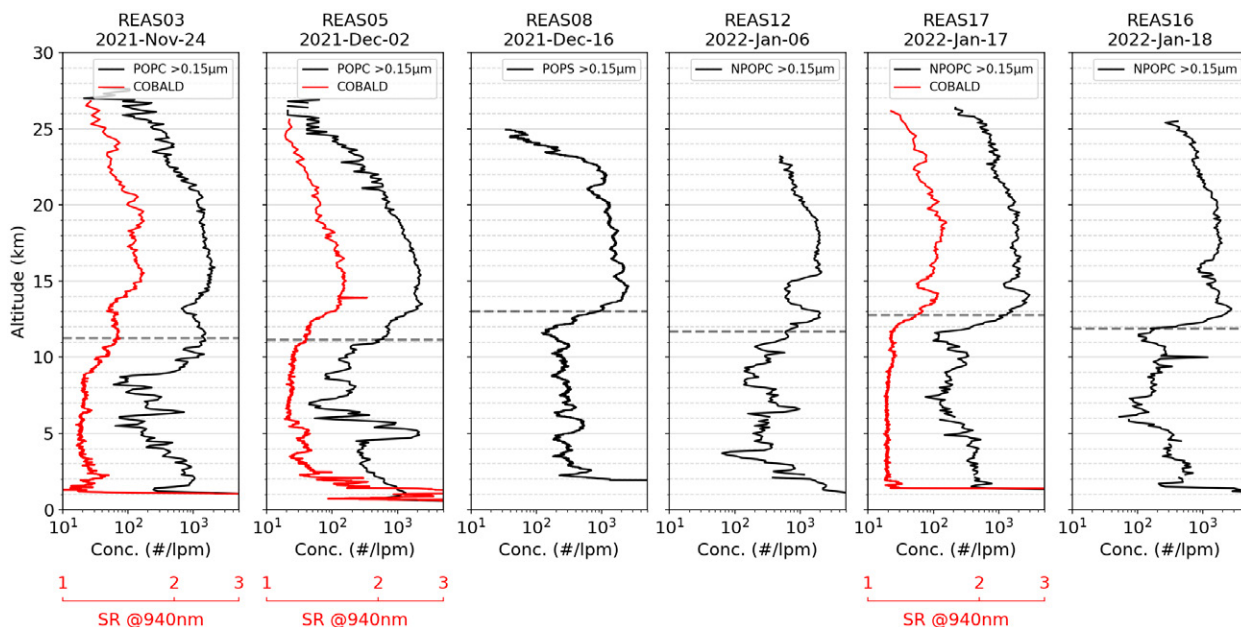


FIG. 8. Aerosol concentration profiles for sizes greater than  $0.15\mu\text{m}$  radius obtained with the Particle Plus Optical Particle Counter (POPC) and the Printed Optical Particle Spectrometer (POPS) during 6 balloon flights of the REAS campaign between November 2021 and January 2022 and corresponding Scattering Ratio at 940 nm for 3 flights of the COBALD instrument coincident with POPC on 11/24/2021, 12/02/2021 and 01/17/2022. Dashed grey lines correspond to tropopause altitude.

measurements integration to further identify stratospheric layers. The presented data corresponds to 2 flights (REAS02 and REAS03) launched on November 24<sup>th</sup>. The POPC profile shown in Figure 9a shows a double peak in the stratospheric aerosol concentration between 9-14 km and 14-21 km. The lower peak is associated with a maximum of CO, CO<sub>2</sub>, and CH<sub>4</sub> concentrations. CO concentration decreases rapidly near the cold point tropopause at 11 km, marking the transition between the troposphere and the stratosphere. For comparison, we used the vertical profile of CO from the Copernicus Atmospheric Monitoring Service (CAMS) monthly mean analysis of November 2021; the levels of CO on November 24<sup>th</sup> have increased by a factor of 1.5 compared to the CAMS model, likely indicating the presence of new sources not accounted for in the model. As we mentioned, a series of pyroCbs were reported during the summer and fall 2021 which impacted the aerosol content in the UTLS. An increase of SR is visible in August 2021 between 40-60N and 9-13 km which seems to persist but diminish by November (Fig. 2d) on the CALIPSO data consistent with OMPS-LP in Figure 7.

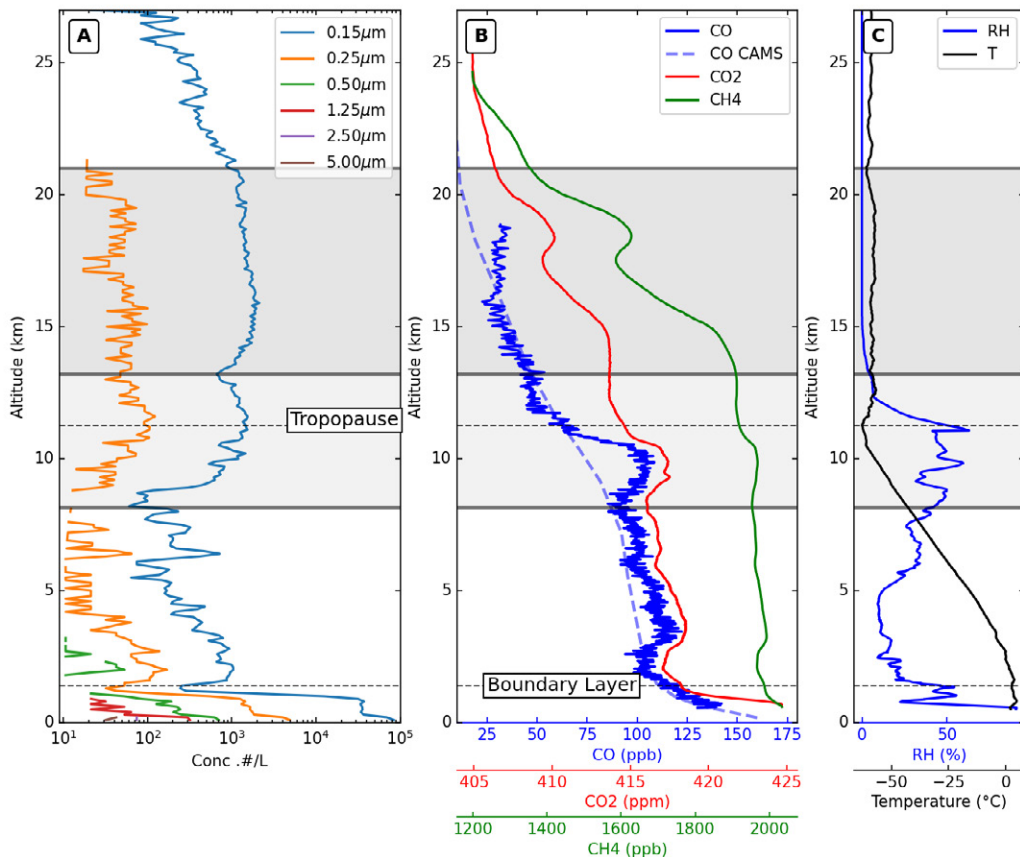


FIG. 9. (a) POPC concentration vertical profiles for different aerosol radii observed on November 24<sup>th</sup> (REAS 3 flight), (b) Aircore CO, CO<sub>2</sub>, and CH<sub>4</sub> vertical concentration profiles for the same date (REAS 2 flight) compared to the CAMS model monthly output for CO, (c) Temperature and Relative Humidity (REAS 3 flight). Dark gray range might have been influenced by La Soufrière eruption whereas light gray range might have been influenced by North America wildfires.

About 100 pyroCb were observed worldwide in 2021 (Fromm et al. 2022) but providing a direct link to a specific event and the increase of CO observed by Aircore is not straightforward. Back-trajectories calculated with HYSPLIT up to 10 days backward could not be matched with any particular emission during 2021 fire season. The KNP complex fire injected smoke up in the UTLS as observed by the CALIOP space-borne LiDAR and was further transported across the Northern

Hemisphere. Together with other pyroCbs observed prior (e.g., Cougar peak fire), it is very likely that the UTLS above Reims was still influenced by those residual smoke plumes in November and influenced our measurements. Our measurements suggest that model simulations miss the influence of wildfires on CO in the UTLS region as well as the radiative and climate impacts of aged smoke layers. Although much progress has been made over the last couple of decades in improving the quality of biomass burning emission inventories like the Global Fire Assimilation System (GFAD) used to drive models like CAMS, large uncertainties remain in the description of the magnitude and injection altitude of wildfires, with expected more significant discrepancies for large fires (Rémy et al. 2017; Pan et al. 2020).

## 6. Conclusion

While it is well established that medium-to-large volcanic eruptions can impact the stratosphere for months to years, the growing influence of wildfires on stratospheric aerosols is an emerging research field. To address this, the REAS project which gathers the expertise of laboratories across Europe and in the United States has been mounted to profile aerosol and trace gas from the ground to the stratosphere onboard light and medium balloon flights. Initiated to respond to sudden events impacting the stratosphere, the REAS 2021-2022 campaign allowed us to test instruments, balloon infrastructure and logistics to be better prepared for the next major stratospheric event. This initiative relies on a unique balloon launch infrastructure installed at the GSMA in Reims since 2018, the only location in Europe to fly medium weight payloads (6 kg) and up to 12 kg through 3 consecutive flights. Although no major event impacted the stratosphere during the campaign, we made measurements within aged volcanic plume of La Soufrière as confirmed by satellite measurements. Aerosol measurements coupled with ozone and greenhouse gases measurements indicated that the UTLS was likely impacted by aged smoke plumes from pyroCbs across North America which took place during the summer and fall 2021. Further studies need to be conducted to better identify potential aerosol and CO sources. Finally, the use of medium-sized balloons allowed us to make the first intercomparisons of instruments dedicated to aerosol measurements as well as making unique stratospheric aerosol collection for subsequent laboratory analysis. Regular balloon flights are now planned from GSMA to continue combining aerosol, O<sub>3</sub> and greenhouse gases measurements to investigate how volcanic eruptions and wildfires affect the stratosphere. For

volcanic study, the measurement of in situ  $\text{SO}_2$  will be investigated following the Morris method (Morris et al. 2010). We are also working on height altitude-controlled balloons to increase aerosol collection time at a predefined altitude for instruments such as the IMPACTOR or the Aerosol sampler. In addition, our teams aim to be ready to respond to the next volcanic and wildfire events that could inject aerosols in the UTLS. However, significant challenges remain such as maintaining teams which can be ready on different sites and improve aerosol forecasting system to localize aerosol plumes. Balloon-borne observations are particularly valuable due to the limited lifetime of satellite missions like with the recent ending of CALIPSO.

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*Data availability statement.* The authors were unable to find a valid data repository for the data used in this study. These data are available from nicolas.dumelie@univ-reims.fr at GSMA UMR 7331, Université de Reims Champagne-Ardenne, Reims, France .

## **7. SIDEBAR**

### *a. GSMA launch facility*

The scientific balloon launch site of GSMA is located on the Moulin de la Housse (MDH) campus in Reims (49.2415°N, 4.0679°E), France. It has a strategic geographical location both in terms of road infrastructure and topography (mainly crop fields), which greatly facilitates the recovery of instruments.

MDH site has been active since 2014 with about 150 successful launches and since 2018, it has been part of the MAGIC initiative (Monitoring Atmospheric composition and Greenhouse gases through multi-Instrument Campaigns) (Crevoisier et al. 2019). Its objective is to provide a facility for the regular launches of light instruments (<3kg) for atmospheric studies such as Aircore atmospheric sampler (Karion et al. 2010; Membrive et al. 2017) and Amulses spectrometers (Joly et al. 2016; Miftah El Khair et al. 2017) for vertical profiles of CO<sub>2</sub>, CH<sub>4</sub>, CO, as well as O<sub>3</sub> ECC probes and aerosol counter such as LOAC (Light Optical Aerosols Counter). The obtained vertical greenhouse gases (GHG) profiles aim to validate atmospheric transport models and to collect

data for comparative studies of satellite measurements (Metop B and C, Sentinel 5-P, OCO-2 or GOSAT) and meteorological models (Joly et al. 2020; Crevoisier et al. 2019).

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