Air quality in the Galapagos Islands: A baseline view from remote sensing and *in situ* measurements

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**Funding information**

Universidad San Francisco de Quito, Grant/Award Numbers: USFQ POA Grant 2018, USFQ Collaboration Grant 2017

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**Abstract**

A characterization of ambient air levels of PM$_{2.5}$, O$_3$, SO$_2$, NO$_2$ and CO in the Galapagos Islands of Ecuador is presented from *in situ* and remote sensing observations. PM$_{2.5}$ was derived from aerosol optical depth (AOD; AERONET) measured at the Universidad San Francisco de Quito, Galapagos Campus (2017–2019). Boundary layer (BL) ozone was obtained from Southern Hemisphere Additional Ozonesondes (SHADOZ) profiles (1998–2016). Background SO$_2$ and pollution events during volcanic eruptions (2005–2018) were estimated from Ozone Monitoring Instrument (OMI) total column measurements through a well-mixed volume approach. Similarly, ambient NO$_2$ was estimated from Ozone Monitoring Instrument (OMI) data (2012–2019). CO was obtained from Measurement Of Pollution In The Troposphere (MOPITT) observations (2012–2017). The study was complemented using Modern-Era Retrospective Analysis for Research and Applications (MERRA-2) reanalysis products and backward trajectory model runs.

From the results, baseline levels of the analysed species (PM$_{2.5}$ = 3.8 μg m$^{-3}$, O$_3$ = 17 ppbv, SO$_2$ = 3.6 ppbv, CO = 80 ppbv, NO$_2$ in populated islands = 23 pptv as one year averages) are comparable with other pristine regions, but some factors can cause increased concentrations. First, high tourism seasons (February–April and July–September) raise background PM$_{2.5}$, NO$_2$ and CO. Furthermore, signals in July–September can be augmented by transport from biomass-burning regions in the Amazon. This latter factor episodically causes ozone to increase up to 45–75 ppbv (1 hr mean). Lastly, volcanic eruptions raise SO$_2$ up to almost 700 ppbv (24 hr mean) and increase PM$_{2.5}$ to 29.5 μg m$^{-3}$ (1 hr mean). The present study provides for the first time baseline levels of air contaminants in the Galapagos, and identifies specific sources whose effect in time is necessary to monitor given global conditions of vulnerable environmental quality.

**KEYWORDS**

AERONET, air quality, criteria pollutants, Galapagos, remote sensing, South America
1 | INTRODUCTION

The Galapagos Islands, which were declared over 40 years ago a Natural Heritage Site for Humanity (UNESCO, 1978), has driven multiple scientific and conservation efforts to study the uniqueness of their species and to oversee their environmental quality. However, limited progress has been achieved with regard to monitoring the Galapagos ambient air, as is evident from the reduced number of available publications on the topic (a brief summary is included below). A main factor that contributes to this gap in information is that continuous air quality measurements, deployed at permanent stations within the archipelago, are still unavailable. Such is the current situation, even though atmospheric monitoring in delicate ecosystems is critical as they are easily affected at very low levels of air contaminants (Stevenson et al., 2000). For this reason, specific legislation in other regions of the world, such as New Zealand or the United States, recommends limits of critical gas concentrations or the imposition of secondary air quality standards to protect wildlife from the adverse effects of air pollutants (Stevenson et al., 2000; US EPA, 2017). Thus, in a global environment where atmospheric composition is changing rapidly, it is of chief importance to monitor the abundance of the main species that potentially influence quality of life in the Galapagos. However, setting up instrumentation that acquires real-time data for a full suite of contaminants to produce high-quality observations in the long term is challenging both financially and logistically. Until such a major effort takes place, the present study characterizes the Galapagos air quality by using a diverse set of best available data including in situ and satellite measurements. Hence, an observation-based study to determine average concentrations of PM2.5, O3, NO2, SO2 and CO that are to be expected in the Galapagos ambient air is presented. In order to place this study in the right context of the current state of knowledge on the subject, a concise summary of previous literature follows.

Sorribas et al. (2015) published a paper on the chemical composition and concentrations of submicron particles and their size distribution measured during a field campaign on San Cristobal in July and August 2011. Contributions from local biogenic and anthropogenic sources were identified, as well as a transported fraction from near the Chilean coast. This particle monitoring effort did not continue beyond the published campaign dates. In addition, Gómez Martín et al. (2013, 2016) studied iodine chemistry and ozone variability in the eastern Pacific marine BL. In these studies, the authors present ground-level ozone measured in the Galapagos during 2000–2002 and 2010–2012, as well as NOx data taken in 2010–2011. These are the only continuous ground-level measurements of ozone and NOx reported for the Galapagos region in the literature, which are discussed further below.

Aside from the aforementioned articles, a source of local information is the Galapagos Report, a publication issued about every two years as a collaborative effort among national and international institutions (Galapagos Conservancy, 2019). For example, Martin et al. (2015) conducted a 30 day environmental monitoring campaign in 2013 on Isabela Island (Galapagos Report, 2013–2014). As part of their study, they performed passive air sampling to measure ambient levels of SOx, NOx, PM, O3 and benzene. They reported not to have found pollution levels of the sampled species. However, they do not provide specific details about the methods used. Also in the Galapagos Report (2015–2016), Orozco et al. (2017) published work on chemical sampling performed on water taxi exhausts between November 2013 and January 2014. From this campaign, they estimated yearly mass emissions of CO, hydrocarbons, NOx3, CO2 and SO2 produced by the water cargo and transport fleet in the town of Puerto Ayora (Santa Cruz Island). Parra (2015) published a set of calculations on the reduction of atmospheric emissions (CO2 and NOx3) at the Universidad San Francisco de Quito (USFQ), Galapagos Campus (San Cristobal), due to the use of solar panels in lieu of the public grid service. Parra documents that the populated islands rely primarily on thermoelectric power generation (there is some wind power on San Cristobal) and emphasizes the need to study further atmospheric emissions. Lastly, Vergara et al. (2005) reported the use of lichens as bio-indicators to detect the presence of atmospheric pollution on the north of Santa Cruz. Their study is qualitative and mainly deals with the solid waste disposal area on the island, which uses open-air combustion.

As presented, there is limited research published on the topic that concerns this study. Particularly, work that compiles in a cohesive manner typical levels of species of interest to air quality in the Galapagos has not been produced to the present. Given the perils that ecosystems are currently facing worldwide (IPBES, 2019), it is of major importance to characterize the Galapagos air quality and identify conditions under which ambient levels of contaminants could increase in the future. With this main motivation in mind, the present study investigates the following research questions:

- What are the current average levels of PM2.5, O3, SO2, NO2 and CO in the Galapagos ambient air?
- What are the scenarios under which some of the analysed species depart from baseline levels leading to pollution episodes in this pristine environment?
To answer the proposed questions, multi-year observations from in situ and remote sensing sources were analysed. The paper is organized as follows. In the Methods Section, data sets, retrievals, processing, and calculations are explained in detail. The Results Section contains proposed average levels for each analysed species along with a discussion of the limitations and comparisons with previous work. Finally, the Conclusions Section states the most relevant lessons learned with respect to this baseline characterization, as well as scenarios under which high levels of contaminants could be encountered.

2 | METHODS

The Galapagos archipelago is a province of Ecuador located 972 km west of the coastline and bounded by coordinates (92.017°W, 1.6°S) and (89.267°W, 1.667°N). Its total surface is 8,010 km² (INOCAR, 2011). The Galapagos Ecuadorian Territory (GET) is depicted in Figure 1. Section 2.1 contains details of the data sets and retrievals used to characterize air quality in the GET; and Section 2.2 explains the data processing to estimate the baseline levels of contaminants.

2.1 | Data

2.1.1 | In situ PM$_{2.5}$ sampling

A brief campaign to sample ambient PM$_{2.5}$ in the Galapagos was executed from July 7 to 21, 2017. During the first week, measurements were taken at the USFQ Galapagos campus on San Cristobal at coordinates 0.895°S, 89.609°W, and 22 masl (Figure 1). The rest of the measurements were taken at a neighbouring station (0.904°S, 89.614°W) run by the Ecuadorian Meteorological Service (INAMHI, Spanish acronym). The sampling site was switched to avoid possible interference from a
work road near the USFQ site. PM$_{2.5}$ observations were taken with two co-located TSI DustTrak II instruments (model 8530) borrowed from the University of North Carolina at Chapel Hill. Real-time signal acquisition was performed with the USFQ’s hardware and software. Briefly, instruments’ signal outputs (0–5 V) were set to be acquired linearly in the 0–200 μg·m$^{-3}$ range at a 1 Hz rate through a National Instruments device. A LabVIEW application was programmed for logging, control and processing. Raw data from each instrument were first reduced to 10 min resolution through a median filter. Measurements from both instruments were in agreement within a 5% difference. Thus, 10 min data from both instruments were averaged. This time series was incorporated into a model to derive PM$_{2.5}$ from remote sensing data.

### 2.1.2 Ozonesondes

The NASA SHADOZ network (Thompson et al., 2004) has launched ozonesondes from the Galapagos since 1998 in co-operation with INAMHI (San Cristobal station; for coordinates, see above). Thus, BL levels of ozone were retrieved from a total of 437 soundings (1998–2016) available from the SHADOZ public archive (https://tropo.gsfc.nasa.gov/shadoz/). Table S1 in the additional supporting information indicates the number of available soundings per year. From the total, eight soundings were filtered out due to invalid data. Additionally, vertical resolution is 100 m for soundings before 2012 and 10 m afterwards. Hence, resolution was homogenized to 100 m for the 429 soundings, which were used in the present study to characterize ambient air ozone.

### 2.1.3 Ground-based remote sensing

The USFQ joined NASA AERONET (Aerosol Robotic Network) (Holben et al., 2001) in July 2017, with one of two monitoring sites at its Galapagos Campus as marked in Figure 1 (the second site is in Quito). Thus, the AOD observations were collected with a CIMEL sun photometer in eight spectral bands (1,640, 1,020, 870, 675, 500, 440, 380 and 340 nm). Measurements were taken with instrument #928, whose head sensor and control box returned to the Goddard Space Flight Center for calibration in August 2018 and again in February 2019 due to a firmware upgrade. Therefore, this work uses level 2 AERONET data with a 3 min temporal resolution from July 21, 2017, to February 11, 2019. This time series was reduced to 1 hr resolution in order to derive ambient PM$_{2.5}$.

### 2.1.4 Satellite remote sensing

**OMI SO$_2$ BASELINE LEVELS**

The ozone monitoring instrument (OMI) measurements (Aura satellite) were used to estimate background and volcanic SO$_2$. Data were obtained from the Goddard Earth Sciences Data and Information Services Center (GES DISC) website. The product used was “OMI/Aura Sulphur Dioxide (SO$_2$) Total Column Daily L2 Global Gridded 0.125°×0.125° V3 (OMSO2G.003)”. All eruptions between 2005 and 2018 were identified from media news. Table 1 shows the volcano eruptions and chosen days for retrievals during episodes of peak SO$_2$. Files were downloaded for the areas that enclose each erupting volcanoes (Figure 1 and Table 1). Background SO$_2$ was estimated for the GET domain using 2016 OMI daily data because there were no volcanic eruptions in that year.

The hierarchical data format - earth observing system version 5 (HDF-EOS5) original files contain daily observations of the SO$_2$ vertical column in Dobson units (DU) for four atmospheric levels: BL, the lower Troposphere (LT) (0–5 km), the middle Troposphere (5–10 km) and the lower Stratosphere (15–20 km) (Krotkov et al., 2016). Files were processed by self-devised MATLAB software. User guidelines recommend retrieving the OMI data for which the solar zenith angle is $<70^\circ$. In addition, it is recommended to use BL data for which the cloud fraction is $<0.3$ (Krotkov et al., 2016). These recommendations were applied by removing data points that did not meet both criteria. Thus, only valid data for both the BL and the LT were averaged over the chosen areas. Total column time series were produced for the event dates in Table 1.

**OMI NO$_2$**

The NO$_2$ data were downloaded from the GES DISC website. The data set used was “OMI/Aura NO$_2$ Total and Tropospheric Column Daily L2 Global Gridded 0.25°×0.25° V3 (OMNO2G.003)”. Background calculations were performed using daily measurements between 2012 and March 2019 for the GET spatial domain. In addition, background NO$_2$ was calculated for two smaller domains that enclose Santa Cruz and San Cristobal islands, bounded by coordinates (90.55° W, 0.77° S, 90.165° W, 0.383° S) and (89.627° W, 0.955° S, 89.23° W, 0.682° W), respectively. This calculation was done because both islands are populated and rely on thermoelectric power generation. The HDF-EOS5 files for the chosen satellite product contain stratospheric and tropospheric NO$_2$ column measurements. Quality flags were applied to remove invalid data. Averages of tropospheric NO$_2$ vertical columns per day were found for the areas of
interest, and these time series were processed to estimate ambient levels of NO2.

**MOPITT CO**

The CO data for the GET region and years 2012–2017 were obtained from the GES DISC Interactive Online Visualization and Analysis Infrastructure (Giovanni) website. The chosen product was MOPITT level 3 monthly mixing ratio profiles (500, 600, 700, 800 and 900 hPa levels) as well as surface mixing ratios (ppbv). The spatial resolution was \(1^\circ\).

### 2.1.5 Modelled data

The lack of in situ measurements in the Galapagos was partially mended by using modelled data from the second Modern-Era Retrospective Analysis for Research and Applications (MERRA-2) (Buchard et al., 2016; 2017). Emphasis was given to PM2.5 and ozone for being the two species for which some observations are available. Retrievals (from GES DISC) were done for the GET spatial domain.

Modelled PM2.5 (mass concentration) was calculated from the following MERRA-2 aerosols products: black carbon (BC), organic carbon (OC), sulfate (S), the 2.5 μm fractions of sea salt (SS25) and dust (D25). This calculation was performed following previous work (Buchard et al., 2016). Hourly data from July 21, 2017, to February 11, 2019 were used to complement the AOD analysis. In addition, monthly MERRA-2 PM2.5 averages (2012–March 2019) were used to help interpret seasonal variability. For ozone, the MERRA-2 monthly profiles (1998–2019) between the surface and 500 hPa were used to complement the SHADOZ analysis.

### 2.2 Estimations of ambient air levels of contaminants

#### 2.2.1 PM2.5

Quantifying ambient air levels of PM2.5 from AOD, using both satellite and AERONET measurements, has been well documented (e.g. van Donkelaar et al., 2006; Schaap et al., 2009; Zhang et al., 2018). In the present study, the AERONET AOD at 500 nm was used to estimate the PM2.5 mass concentrations through simple linear models.

First, an experimental model was built by finding a correlation between the PM2.5 measurements (July 7–21, 2017) and AOD. Owing to logistical reasons, the observations did not occur simultaneously for the AERONET measurements began on July 21, 2017. Thus, a statistical

| Island | Volcano | Height (masl) | Coordinates of the bounded areas | Area (km²) | Eruption events |
|---|---|---|---|---|---|
| Isabela | Cerro Azul | 1,640 | \(-0.790^\circ, -1.030^\circ\) | 960 | May 30–June 10, 2008 |
| | Wolf | 1,710 | \(-0.200^\circ, -0.200^\circ\) | 960 | May 25–June 2, 2015 |
| Fernandina | La Cumbre | 1,476 | \(-0.980^\circ, -1.290^\circ\) | 1,687 | June 26–July 4, 2018 |
| | Sierra Negra | 1,124 | \(-0.690^\circ, -0.790^\circ\) | 960 | May 13–18, 2005 |
| | Cataratas | 1,590 | \(-0.533^\circ, -0.730^\circ\) | 960 | October, 22–30, 2005 |
| | Cerro Azul | 1,640 | \(-0.200^\circ, -0.790^\circ\) | 960 | June 16–22, 2018 |
| | Wolf | 1,710 | \(-0.200^\circ, -0.790^\circ\) | 960 | May 30–June 10, 2008 |

*Table 1: Volcano information, coordinates and surface of the bounded areas around each volcano. Dates correspond to event days for which ambient air SO2 mixing ratios were calculated.*
strategy was designed to find the best possible correlation between the daily trends of the PM$_{2.5}$ and AOD during the same hours of the day. To this end, an equal number of days for the AOD observations, taken right after the PM$_{2.5}$ campaign, were used (July 21–August 7). Only daylight hours 0600–1800 LST (UTC-6) were compared for it being the time when the sun photometer acquired valid data. To find trends, median diurnal variation (MDV) curves were built for both quantities. The MDV for PM$_{2.5}$ was found by overlapping 10 min data points in a 24 hr plot and finding the median within 10 min bins (see Figure S1 in the additional supporting information). Similarly, an MDV trend was built with 3 min AOD data. Both MDV curves were interpolated to the same time stamp (10 min) and a best-fit linear regression was found.

The experimental model was obtained when the AOD happened to range from 0.050 to 0.103. However, the entire AERONET time series has a seasonally higher AOD and a high aerosol event in summer 2018. Therefore, an alternative model was built in order to better suit the higher AOD. Hence, different correlation trials were performed between the MERRA-2 PM$_{2.5}$ and the AOD observations (2017–2019, 1 hr resolution). A best-fit regression was found for AOD > 0.2. Thus, the slope of the experimental model was used to scale AOD ≤ 0.2, while the alternative model was used for AOD > 0.2. The two linear models are presented in the Results Section, along with the 2017–2019 AOD time series scaled to PM$_{2.5}$ mass concentrations. Uncertainty in the proposed PM$_{2.5}$ characterization was estimated by finding the bias and standard deviation of the error in the models (Brock and Richardson, 2001).

Variability of the PM$_{2.5}$ within a year was explored further with monthly MERRA-2 PM$_{2.5}$ data (2012–2019). Finally, the high aerosol event in 2018 was studied with the help of the hybrid single-particle lagrangian integrated trajectory (HYSPLIT) model (Stein et al., 2015). This analysis was complemented using Moderate-resolution Imaging Spectroradiometer (MODIS) images (https://worldview.earthdata.nasa.gov) (data not included).

### 2.2.2 | Ozone

The SHADOZ soundings were treated statistically within the first 5 km by obtaining profiles for the 5th, 50th and 95th percentiles. Furthermore, seasonal distributions of ozone mixing ratios (0–2 and 2–5 km) were investigated through histograms. Distinctive distribution modes were analysed for the warm wet season (WWS), which runs from December to May, and the dry cool season (DCS), from June to November. The SHADOZ data have several gaps due to inconsistent sampling frequency with several months and years with no soundings. To compensate for this lack of in situ data, modelled ozone profiles from the MERRA-2 reanalysis were used to interpret better ozone seasonal variability. In addition, events of high ozone in the SHADOZ profiles were studied in light of HYSPLIT model-backward trajectories. Satellite images helped track pollution from biomass-burning regions over the Amazon, but were not included. The influence of El Niño and La Niña (EN/LN) conditions were investigated in time series beginning in 1998 (SHADOZ and MERRA-2). To this end, data were classified according to Oceanic El Niño Index (ONI) ranges (NOAA, 2019). Table S2 in the additional supporting information includes definitions and the ONI categories. Briefly, very strong EN events are associated with an ONI > 2, “no event” is considered for −0.5 < ONI < 0.5, and a strong LN is given by −2.0 < ONI < −1.5.

#### 2.2.3 | SO$_2$

In order to estimate ambient air SO$_2$ (mixing ratios) from satellite total column measurements, a well-mixed volume model of the atmosphere was applied. The depth of the column used for the well-mixed approximation was either the BL’s or the LT’s, depending on the satellite product used. The steps taken in this calculation were the following:

1. The Dobson unit definition (1 DU = 0.001 cm of pure gas at 273 K and 1 atm) was applied to find the thickness (z) of the pure SO$_2$ layer that corresponds to satellite column observations in the BL and the LT.

2. The SO$_2$ column burden (n) (molecules cm$^{-2}$) was obtained from Equation (1); where $N_A$ is Avogadro’s number; $R$ is the ideal gas law constant; $T = 273$ K and $P = 1$ atm:

   \[
   z = \frac{n}{N_A} \left( \frac{RT}{P} \right)
   \]  

3. The total number of SO$_2$ moles was estimated from satellite column measurements (BL or LT), the selected area around each volcano (Table 1 or the GET for background calculations), and the compound’s molecular weight.

4. Mean vertical pressure and temperature profiles over the Galapagos were obtained by averaging all the SHADOZ pressure and temperature profiles (see Figure S2 in the additional supporting information). Thus, an average vertical profile for air density was determined.

5. An average depth of 1.5 km was used for the BL, while 5 km was used for measurements in the LT. Each
column was divided into 100 m slabs. The number of air moles was determined with the slab’s volume and air density at the corresponding pressure level in the centre of the slab.

6. The total number of air moles in the atmospheric column of interest was found by integrating all the column slabs.

7. Finally, the SO$_2$ mixing ratio (ppbv) was found from the ratio of the number of SO$_2$ moles to the number of air moles and applying the $10^9$ factor.

Uncertainty in the above calculation was estimated in terms of the error caused by assuming a specific depth for the BL. This aspect was investigated by finding ambient SO$_2$ with different BL depths and evaluating the percentage error induced. Finally, the results were discussed in light of concentrations measured in other pristine regions.

### 2.2.4 | NO$_2$

Ambient air NO$_2$ was calculated with the same above method used for SO$_2$. This approach was adopted in lieu of more complex methodologies that use ground measurements or NO$_2$ vertical profiles (unavailable in the present case) to find scaling factors to be used with satellite observations (e.g. Anand and Monks, 2017). Hence, estimates of background NO$_2$ mixing ratios were obtained with Steps 3–7 described above (OMI NO$_2$ column measurements are given in molecules cm$^{-2}$), although the column depth used in this calculation was 10 km. Monthly averages from multi-year data were overlapped to find an annual trend. These estimations are discussed in terms of observed ranges measured in 2010–2011 by Gómez Martín et al. (2016).

### 2.2.5 | CO

Ambient levels of CO for the study area were directly downloaded as mixing ratios (MOPITT). Multi-year monthly time series were overlapped to investigate seasonal distribution and variability. CO vertical mixing ratios for all years were averaged and plotted as a colour map to help better visualize the data.

### 3 | RESULTS

#### 3.1 | PM$_{2.5}$

The experimental linear model found by correlating the PM$_{2.5}$ observations with the AOD is presented in Figure 2a. The slope of this model, whose value is 41, was used as a scaling factor to convert most AOD measurements (Figure 2b) into the PM$_{2.5}$ mass concentrations (> 90% of the data). However, the correlation coefficient for the empirical linear model is low ($R^2 = 0.21$), which is attributed to several factors. First, measurements of both quantities were not taken simultaneously, although the AOD observations were taken immediately after the PM$_{2.5}$ campaign and within the same area. In addition, the PM$_{2.5}$ time series is short. Consequently, an argument can be made that PM$_{2.5}$ sampling over a couple of weeks is not representative of yearly variability. Furthermore, AOD is a total column measurement, whereas PM$_{2.5}$ is an ambient air measurement. From this perspective, there is some aerosol load within the atmospheric column that is not necessarily distributed within the BL. To partially compensate for these limitations, a comparison was performed between the MERRA-2 PM$_{2.5}$ and the AOD observations (2017–2019, 1 hr resolution). When overlapped, the bulk of both data sets roughly follow the same major features (see Figure S3 in the additional supporting information). Thus, correlations were investigated among the two time series for different months, seasons and AOD ranges. Data sorted for the AOD $> 0.2$ yielded a linear regression with a correlation factor $R^2 = 0.45$ and a slope of 50 (Figure 2c). These data comprise seasonally increased AOD as well as a distinctive pollution event in summer 2018 (these features are analysed in detail below). Other correlation trials yielded poor comparisons for which they were not included. Thus, the experimental scaling factor for AOD $\leq 0.2$ was retained and the modified scaling factor when AOD $> 0.2$ was incorporated. With this improvement, the PM$_{2.5}$ mass concentrations from July 21, 2017, to February 11, 2019 (1 hr data and monthly means) were estimated (Figure 2d). Therefore, the PM$_{2.5}$ annual mean is 3.8 μg·m$^{-3}$. As a reference, the World Health Organization (WHO) (2006) Air Quality Guidelines (AQG) recommend 10 μg·m$^{-3}$ of PM$_{2.5}$ as an annual mean to protect public health.

Figure 2a,c depict the bias and the standard deviation of the error (stde) in the two linear models. Hence, uncertainty (within 1 stde) in calculations presented in Figure 2d, for data points $< 10$ μg·m$^{-3}$ is 14%, while for higher values uncertainty is 43%. From a practical standpoint, these results can be summarized in two ways: (1) the only measurements currently available in the Galapagos from which ambient PM$_{2.5}$ can be estimated are the AOD observations (USFQ site); and (2) from the best available information, the factor to scale AOD into the PM$_{2.5}$ is 41–50, with a larger uncertainty at higher AODs. Additional studies that gather extended in situ measurements are needed in future studies to refine or modify the proposed scaling method.
With respect to seasonal variability, Figure 2b indicates that within a year, the AOD increases from February to April and later again between July and September. The origin of these signals was investigated using the individual aerosol contributions to the MERRA-2 PM$_{2.5}$. Aside from the sea salt contribution, which is the largest, there is an increased OC signal from February to April and from July to September–October (see Figure S4 in the additional supporting information). Also, the sulfate signal is higher from January to April, and there are distinctive peaks on June 29 and July 24, 2018. Several circumstances are consistent with individual aerosol contributions. First, time periods for the increased PM$_{2.5}$ coincide with high tourism seasons, as can be inferred from national statistics (DPNG, 2018). In particular, February–April are the preferred months for warmth. These months overlap with spring break vacation in the United States, while July–September corresponds to the summer vacation in continental Ecuador and the Northern Hemisphere. A check done with the MERRA-2 monthly data (2012–2019) is consistent, in particular with the March peak. Signals associated with increased tourism activities on the islands are also evident in other trace gases, as will be discussed below. A second factor involves transport during the biomass-burning season over the Amazon, which occurs in July–November (Gonzalez-Alonso et al., 2019). These events have an impact as air masses move over the Andes and into the Galapagos region due to the prevalence of the easterly synoptic flow. Lastly, volcanic eruptions within the archipelago impact the AOD and its associated PM$_{2.5}$. An example is discussed below.

A high aerosol event on June 29, 2018, is evident in the AERONET data as AOD reached 0.54 with a second peak of 0.58 on July 24, 2018 (Figure 2b). These episodes are mainly associated with the eruption of Sierra Negra volcano on Isabela Island as well as with transported air masses at higher altitudes from continental South America. Sierra Negra erupted on June 26, 2018, with its activity lasting until August 23, 2018 (IG, 2018). MODIS satellite images (data not included) show the presence of a thermal anomaly on Sierra Negra volcano (overpass on June 27), surrounded by areas of a high AOD in subsequent days. Backward trajectories for June 29 (see Figure S5 in the additional supporting information) indicate that air masses at 1,500 m recirculated from over Isabela Island. At the 5,000 m level, air masses originated from biomass-burning regions in the Brazilian Amazon, where at the time there was also a large transport event arriving from Africa. Aerosols distributed within the atmospheric column, whose origin is transport from volcanic sources or biomass-burning regions, can be detected by the sun photometer on San Cristobal. However, their true impact on air quality would only be
measured by a PM$_{2.5}$ ground sensor deployed within the archipelago (currently unavailable). At the moment, Figure 2d reports a maximum ambient PM$_{2.5}$ of 29.5 μg·m$^{-3}$ (1 hr average) for the episode in summer 2018, but future studies need to incorporate PM$_{2.5}$ ground measurements to decouple the aerosol load within the BL from the entire atmospheric column.

As presented, baseline levels of PM$_{2.5}$ in the Galapagos were determined as well as distinctive sources that cause increased signals. The calculated mean concentration of 3.8 μg·m$^{-3}$ is an annual average, which is comparable with observed values in other remote regions such as Hawai‘i (3.7–5.4 μg·m$^{-3}$, 24 hr mean) (Department of Health, 2016), but lower than levels on the Canary Islands (15.2–16.4 μg·m$^{-3}$, annual mean) (López-Villarrubia et al., 2010). Furthermore, van Donkelaar et al. (2006) proposed average values of AOD and their corresponding PM$_{2.5}$ for the entire globe between 2001 and 2002, which over the Galapagos region is about 0.2 and 10 μg·m$^{-3}$, respectively. As an additional check, in the Galapagos environment AOD and its associated PM$_{2.5}$ should be significantly lower than those over populated continental areas. For example, empirical scaling factors inferred from studies in regions such as the Netherlands (Schaap et al., 2009) are about 60, whereas over polluted urban areas factors are 70–90 or higher (Zhang et al., 2018). Therefore, the proposed scaling methodology yields ambient PM$_{2.5}$ in the Galapagos expected for a pristine environment.

3.2 | Ozone

Ozone mixing ratios (SHADOZ) in the first 5 km above San Cristobal are depicted in Figure 3a. From the bulk of the data, ozone levels within the first 2 km are usually < 35 ppbv, as indicated by the 5th and 95th percentile profiles. For practical purposes, it is assumed that individual data points are representative of 1 hr averages because readings were taken in a pristine region where background ozone levels vary slowly. Thus, an average of all points in the BL (< 2 km) can be interpreted as an annual mean (17 ppbv). Typical values ranged from 10 to 20 ppbv, as given by the 50th percentile. These near-surface levels are consistent with Gómez Martin et al. (2016) who present surface ozone data in similar ranges collected continuously with an ultraviolet monitor between 2000 and 2002 on San Cristobal and between 2010 and 2012 on Isabela. Thus, average mixing ratios of ozone are below the 40 ppbv threshold established to calculate critical levels above which there is damage to ecosystems (Stevenson et al., 2000). As an additional reference, the recommended WHO AQG for ozone is 50.9 ppbv as an 8 hr mean (100 μg·m$^{-3}$ in standard conditions).

With regard to seasonal variability, lower levels of ozone (0–2 km) are typical during the WWS, as the mean ozone mixing ratio within 1 SD (standard deviation) is 15.6 ± 6.3 ppbv (see the histograms in Figure S6 in the additional supporting information). In contrast, higher levels of ozone are more frequent during the DCS, when the mean mixing ratio is 19.2 ± 5.9 ppbv. At higher altitudes (2–5 km), ozone mixing ratios are higher in both seasons: 25.7 ± 7.5 versus 33.4 ± 11.8 ppbv for WWS and DCS, respectively. A check performed with MERRA-2 profiles reveals similar seasonal distributions of ozone (0–2 and 2–5 km), although modelled data are generally higher (7 ppbv, on average) than the SHADOZ observations in both seasons (see Figure S7 in the additional supporting information). Increased levels of ozone at higher altitudes during the DCS are consistent with the transport of polluted air masses during biomass-burning months in the Amazon, as concise examples (discussed below) demonstrate.

Percentile profiles in Figure 3a were calculated excluding four soundings that have ozone levels out of the range of the data bulk, because their mixing ratios within 0–5 km are 70–107 ppbv. These profiles correspond to about 1% of the data (4/429), and flights were launched on September 16 and 30, 2004, October 4, 2007, and August 23, 2012 (Figure 3b). Of this subset of soundings, the profile on August 23, 2012, shows 45 ppbv at the surface and 75 ppbv at 1.5 km. Such ozone mixing ratios in the BL are not expected in a pristine environment, and they are rather comparable with those found over urban areas (Cazorla, 2016). Puerto Baquerizo Moreno, the town on San Cristobal from which ozonesondes were launched, has a population of about 6,600 people according to the 2010 Census (INEC, 2010). There is light road traffic as well as plane and maritime traffic associated with tourism and daily citizen activities. Only 1% of data having higher than expected ozone levels near the surface is an indicator that local activities are not the cause for such mixing ratios, but rather that air quality in the Galapagos can be affected by advection of pollution (e.g. biomass-burning regions are sources of ozone precursors such as CO and NO$_x$).

Transport of polluted air masses is evident in 10 day backward trajectories (HYSDPLIT) for the four high-ozone soundings (see Figures S8 and S9 in the additional supporting information). The two profiles on September 16 and 30, 2004, are associated with intense fires in the Amazon which are evident in satellite imagery (data not included). On October 4, 2007, levels of ozone at 3 km over San Cristobal were as high as 72 ppbv. The backward trajectory indicates that air masses at 3 km
originated at 6.5 km from a region of intense fires over northeast Brazil. The pollution event on August 23, 2012, is remarkable from the perspective that pollution from two biomass-burning regions in the Amazon and Africa arrived simultaneously over the Galápagos. On this particular day, ozone levels above San Cristóbal at 1.5 and 3.0 km were 75 and 90 ppbv and originated at 5.5 and 6.5 km over central Brazil and to the east, over the Atlantic, respectively. At the same time, air masses at 5 km had 107 ppbv of ozone and were transported from about 8 km over West Africa. The MODIS images for the 10 day backward trajectories indicate intense biomass-burning activity in both source regions. These observations are consistent with previous studies that document a usual pattern of pollution transport over the Galápagos from the north of Brazil, mainly during the August–October season (Oltmans et al., 2001; Thompson et al., 2010).

The possible effects of EL/LN conditions on surface ozone were examined according to the ONI categories (see Table S2 in the additional supporting information). There are 12 SHADOZ profiles that were launched in years with very strong EN conditions and 109 during weak to medium EN intensities. On the other hand, 81 soundings were launched in years with “no event”, 88 during weak to medium LN, and 139 during strong LN conditions. The four high-ozone episodes discussed above were detected during “no event” years (2004, 2012) and a strong LN year (2007). From a physical perspective, under EN conditions, easterlies around the Equator weaken, which should decrease the advection of air masses loaded with precursors from continental South America. Thus, high-ozone episodes should be more frequently observed under normal easterly conditions, provided there is biomass burning upwind, as is actually the case for the four detected soundings. However, there are very few SHADOZ samples with high ozone to provide sufficient evidence. As per the MERRA-2 profiles, they lack the high-ozone structure detected episodically in the SHADOZ soundings. As a result, data distributions in the BL for different ONI ranges in both data sets overlap, yielding no difference (see Figure S10 in the additional supporting information). Thus, from the SHADOZ observations, there is some indication that episodes of transport-associated high ozone in the Galápagos occur under conditions unrelated to EN. However, further investigation is needed with additional sampling and validated models to better understand the EN/LN effects in surface ozone.

From the above discussion, the main influence that has a clear impact on the baseline levels of ozone in the Galápagos is the advection of air masses rich in ozone precursors from biomass-burning regions in the Amazon. This factor was found to increase ozone mixing ratios in the BL to a detected range between 45 and 75 ppbv (1 hr data) (Figure 3b). Statistically, profiles with high ozone correspond to 1% of the available soundings, but this percentage does not indicate the true frequency of high-ozone events as sampling in many months and years is scarce (see Table S1 in the additional supporting information). Therefore, the real frequency and magnitude of such events need further investigation. Unfortunately, continuous ground-level ozone monitoring is unavailable and the SHADOZ soundings have not been launched since 2016 (only one flight was launched that year). Thus, permanent ozone monitoring and more intensive upper air studies are needed to identify and track the seasons.
and magnitudes of ozone pollution episodes in the Galapagos.

### 3.3 Background and volcanic SO₂

As explained above, a simple well-mixed volume model that uses 1.5 and 5.0 km for the depth of the BL and LT, respectively, was applied to determine the SO₂ mixing ratios for both background levels and high-pollution events due to volcanic eruptions.

Background levels of SO₂ for the GET domain were estimated using the 2016 OMI data for being a year free of volcanic eruptions. Figure S11 in the additional supporting information contains the BL column measurements retrieved from OMI, with their corresponding SO₂ mixing ratios. From these calculations, SO₂ in the BL is 3.6 ppbv, as an annual average, with the bulk of measurements (> 85%) between 0.5 and 6.0 ppbv. These results resemble the 0–5 ppbv annual average (2011–2015) on the Hawaiian Islands, as reported by the Department of Health (2016). For reference, the critical level (annual mean) for the SO₂ is 7.6 ppbv (20 μg m⁻³ in standard conditions). The WHO AQG is 7.6 ppbv as a 24 hr mean.

With respect to episodes of high levels of SO₂, the main sources are volcanic eruptions that mainly occur on the youngest islands, Fernandina and Isabela (Figure 1 and Table 1). Calculated SO₂ mixing ratios (24 hr averages) in the ambient air, during eruptions of active volcanoes La Cumbre on Fernandina and Sierra Negra, Cerro Azul and Wolf on Isabela, are presented in Figure 4a,b, respectively.

The volcano that has erupted more frequently in the past years is La Cumbre, which is the youngest of the four volcanoes. The May 2005 eruption (Figure 4a) caused atmospheric SO₂ levels to increase to 321.7 ppbv on May 13 in the LT and to 394 ppbv in the BL on the next day, which largely exceeds critical levels. La Cumbre volcano is located on an uninhabited island for which there was no harm to human health. It was reported that the impact on vegetation and wildlife due to lava flows was minimal (Global Volcanism Program, 2005), but there is no specific reference to the impact of high ambient SO₂. The following eruption occurred in April 2009, when SO₂ levels in the BL escalated from 17 ppbv on April 11 to 359 ppbv on April 19. The eruption plume drifted west of Fernandina, over the ocean, as indicated by satellite images (data not shown). Eruptions in 2017 and 2018 caused SO₂ levels in the ambient air to increase to 27 ppbv, which is significantly lower than the 2005 and 2009 levels, although it is five times higher than background mixing ratios.

With regard to eruptions on Isabela Island, Sierra Negra’s 2005 eruption (Figure 4b) caused an estimated SO₂ pollution event of 2,662 ppbv in the LT (October 23), the highest of all recent eruptions. With regard to the fate of polluted air masses, OMI images (data not shown) indicate that SO₂ plumes headed west, which means the inhabited areas east of the volcano were not directly affected. Sierra Negra volcano also erupted recently, from June 26 to August 23, 2018 (Figure 4b shows the first nine days of activity). The peak SO₂ levels in the BL reached 387 ppbv on July 3, comparable with the peak 3 hr average of 472 ppbv that the eruption in the Hilo volcano induced in January 2015 in Hawai’i (Department of Health, 2016). From previous discussions, this plume induced increased PM₂.⁵ away from the eruption (San Cristobal), but there is no SO₂ ground sensor within the archipelago to have a reference from in situ measurements.

Also on Isabela Island, Cerro Azul volcano erupted in May–June 2008, which produced ambient SO₂ near 100 ppbv. Finally, Wolf volcano caused major commotion due to its spectacular 2015 eruption that caught the attention of both scientist and tourists, and merited large media coverage. On May 25, low tropospheric SO₂ levels rose to 230 ppbv. On May 27, ambient air levels of SO₂ reached 184 ppbv. OMI images from May 25–26 (data not shown) indicate that the SO₂ plume spread over most of the Galapagos. Therefore, it is logical to conclude that this particular eruption did have a direct impact on the Galapagos air quality, although no further information was found from local institutions or the media.

A main source of uncertainty in the proposed estimations is the assumption of a fixed depth for the BL (1.5 km). In order to investigate the effect of this dilution factor, calculations of ambient SO₂ were performed when the depth of the BL is 1 and 2 km (see Figure S12 in the additional supporting information). For example, during the 2005 eruption of Sierra Negra Volcano, the reported ambient SO₂ on October 28 is 695 ppbv. However, at BL depths of 1 and 2 km, calculated mixing ratios are 1,016 and 535 ppbv, respectively. For all cases, the proportionality in error remains constant because the dilution factors are constant. From these calculations, mixing ratios are roughly 23% lower if the BL is deeper than 1.5 km and 46% higher if it is shallower. Given the lack of ground measurements, at the moment it is only possible to offer a gross estimate of error. However, these estimations need to be investigated in more depth as ground-based measurements become available in the future.

### 3.4 NO₂

As the Galapagos archipelago is located in a remote region in the Pacific Ocean, it is reasonable to assume
that ambient air NO$_2$ is that of the well-mixed clean background atmosphere. The monthly mean time series for NO$_2$ mixing ratios (from OMI data, 2012–2019) in the GET region is depicted in Figure 5a (blue line). Thus, NO$_2$ is 12 pptv as an annual average, but levels slightly rise around March and August due to the high tourism seasons. The same background estimations were performed for areas that enclose Santa Cruz and San Cristóbal Islands, for being populated and for mostly relying on thermoelectric power generation. Average baseline NO$_2$ mixing ratios on these two islands (Figure 5a, orange line) almost double the background GET levels, as the annual mean concentration is 23 pptv. Compared with other regions, these mixing ratios are lower than, for example, the 24 hr annual mean in Hawai’i (2–4 ppbv) (Department of Health, 2016). However, NO$_2$ measurements performed with a chemiluminescence instrument by Gómez Martín et al. (2016) in the Galapagos show that during mid-morning and into the early afternoon, NO$_2$ levels are under the detection limit of the instrument, while there usually is a detectable morning peak of about 100 pptv. Thus, calculations performed in the present study are in agreement with the order of magnitude of in situ measurements detected by Gómez Martín et al. Finally, estimated baseline mixing ratios are well below the critical level for NO$_x$ (Stevenson et al., 2000), which for NO$_2$ correspond to an annual mean of 15.9 ppbv (30 μg m$^{-3}$ in standard conditions). The AQG for NO$_2$ (WHO) is 53 ppbv as an 8 hr mean (100 μg m$^{-3}$ in standard conditions).

### 3.5 CO

CO near the surface from MOPITT observations (monthly averages, 2012–2017) is presented as a time series in Figure 5b. There is a seasonal pattern of CO variability that consistently repeats among all years as levels increase in March–April, although a significantly larger peak is evident in August–September. The enhancement of the latter peak is consistent with high signals due to advection from biomass-burning regions. This signal is evident in the vertical structure of CO presented in a colour map in Figure S13 in the additional supporting information, where higher levels in July–September extend up to 500 hPa. This pattern coincides with the PM$_{2.5}$ and NO$_2$ signals during similar time periods. Deeter et al. (2013) validated CO MOPITT measurements in several points around the globe. From the present study, it can be inferred that the standard deviation of the error in the MOPITT measurements is about 16%. However, there are no local measurements to have a more accurate reference for ground-level CO. From the monthly CO in Figure 5b, the typical range is 66–117 ppbv and the annual mean is 80 ppbv. Compared with
other regions, CO in the Galapagos is lower than, for example, in Hawai‘i (600–700 ppbv, annual mean) (Department of Health, 2016). For reference, the American air quality standard for CO is 9 ppm (8 hr mean) (US EPA, 2017), while there is no recommended WHO AQG nor critical level for this contaminant.

3.6 | Summary of baseline levels

Baseline concentrations of atmospheric constituents relevant to air quality in the Galapagos are presented in Figure 6 as time averages and typical ranges from multi-year data. Departures from baseline levels are presented as maxima from each data set. Increased tourism activities around February–March and July–September are evident in the PM2.5, NO2 and CO signals. According to national statistics, tourism in the Galapagos has grown at a compound yearly rate of 6.71% in the last 30 years (DPPNG, 2018). Thus, it is necessary to monitor in time the impact of this source along with the biomass-burning season in the Amazon that overlaps with the July–September period. Transport due to the latter factor is capable of inducing high ambient ozone. Volcanic eruptions also can affect ambient air by raising SO2 and PM2.5.

FIGURE 6  Baseline levels (Avg.) of species relevant to air quality in the Galapagos Islands, Ecuador, as one year averages. Data sources: AERONET AOD (2017–2019) for deriving PM2.5; SHADOZ profiles (1998–2016) for ozone; OMI measurements for background and volcanic SO2 (2005–2018); MOPITT observations (2012–2017) for ambient CO; OMI NO2 (2017–2019) was used for the entire Galapagos Ecuadorian Territory (GET) and two populated islands (PI). Average times for typical ranges and maximum detected values (Max.) are indicated in parentheses.

4 | CONCLUSIONS

The Galapagos Islands currently lack an air quality monitoring network. In spite of this limitation, ambient levels of species relevant to air quality were characterized from in situ and remote sensing observations. On average, baseline levels of the analysed species correspond to concentrations expected in the clean background atmosphere. However, specific conditions related to tourism, location of the islands downwind of continental South America and the volcanic nature of the archipelago have an impact on air quality. Baseline levels of PM2.5, O3, SO2, NO2 and CO range (as monthly averages) at about 2.4–6.7 μg·m⁻³, 10–20 ppbv, 0.5–6.0 ppbv, 18–27 pptv (populated islands) and 66–117 ppbv, respectively. Tourism intensifies around February–April and July–September, which raises the background PM2.5, NO2 and CO. These signals can be augmented by advection during the biomass-burning season in the Amazon, which overlaps with the July–September months. This factor was found to episodically induce high ozone (45–75 ppbv, 1 hr mean) in the boundary layer. Four high-ozone profiles were encountered in the 1998–2016 SHADOZ data set, but the frequency and intensity of these events need further investigation as several months and years have no samples. Events of volcanic eruptions also impact air quality by raising the ambient SO2 to almost 700 ppbv (24 hr mean). A recent volcanic eruption (2018) on Isabela impacted the PM2.5 on San Cristobal by raising levels to 29.5 μg·m⁻³ (1 hr mean). At the moment, the AERONET sun photometer at the Universidad San Francisco de Quito (USFQ), Galapagos Campus (San Cristobal), is the only instrument from which PM2.5 can be estimated with the scaling method proposed in this study. Given the current regime of increasing global pollution, in situ monitoring of air quality in the Galapagos is needed, particularly to track departures from baseline levels caused by the identified sources of contaminants. The characterization presented in this study is a contribution towards advancing this endeavour.

ACKNOWLEDGEMENTS

This study was funded by USFQ Collaboration Grant 2017 and POA Grant 2018. Thanks for the support of the Galapagos Science Center. Thanks to Brent Holben for supporting the integration of USFQ to AERONET and to David Giles and Mikhail Sorokin for AERONET data production and support. Luis Tasipanta (USFQ) overlooks our Galapagos instrument. Thanks to William Vizuete (UNC Chapel Hill) who lent the DustTrak instruments. Thanks to Anne Thompson (NASA), Bryan Johnson (NOAA) and INAMHI for SHADOZ soundings. Thanks to INAMHI and Julieta Juncosa (USFQ) for help with the
Galápagos campaign. Thanks to both reviewers of this paper for thorough feedback. There is no conflict of interest.

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SUPPORTING INFORMATION

Additional supporting information may be found online in the Supporting Information section at the end of this article.

How to cite this article: Cazorla M, Herrera E. Air quality in the Galapagos Islands: A baseline view from remote sensing and in situ measurements. *Meteorol Appl*. 2020;27:e1878. https://doi.org/10.1002/met.1878