ABSTRACT

We investigated the variability of the aerosol scattering ($\sigma_{\text{sp}}$; 1974–2015) and absorption ($\sigma_{\text{ap}}$; 2000–2015) coefficients at the Mauna Loa Observatory using in situ measurements. Although $\sigma_{\text{ap}}$ decreased during the morning (1.85 ± 3.43 Mm$^{-1}$ at 550 nm, 8–11 local standard time [LST]), it increased during the afternoon (3.72 ± 7.63 Mm$^{-1}$ at 550 nm, 14–17 LST) due to the development of thermally induced boundary layer winds. No distinct diurnal variation was observed in $\sigma_{\text{sp}}$. The obvious increase in $\sigma_{\text{ap}}$ and $\sigma_{\text{sp}}$ during the spring under free troposphere conditions (8–11 LST) is attributed to long-range-transported aerosols from Asia, especially dust and pollution aerosols from Northeast Asia and biomass burning aerosols from Southeast Asia. Accordingly, $\sigma_{\text{ap}}$ increased from 1974 till 2015 (at 1.89% year$^{-1}$), whereas no significant trend was noted for either $\sigma_{\text{sp}}$ or $\sigma_{\text{ap}}$ from 2000 till 2015. An increasing trend for $\sigma_{\text{ap}}$ prevailed in air masses originating in Northeast Asia (+0.51 Mm$^{-1}$ decade$^{-1}$).

Keywords: Aerosol in situ measurement; Aerosol scattering coefficient; Aerosol absorption coefficient; Mauna Loa.

INTRODUCTION

Optical and radiative properties of atmospheric aerosols depend on their chemical compositions, shapes, and particle size distributions (Haywood and Ramaswamy, 1998; Delene and Ogren, 2002; Jacobson, 2002). These properties exhibit high spatial and temporal variations because of the relatively short lifetime and uneven geographical distribution related to emissions, chemical processes in the atmosphere, and weather patterns (Delene and Ogren, 2002; Andrews et al., 2011; Boucher et al., 2013; Collaud Coen et al., 2013; Park et al., 2019). Even though the space-based and ground-based remote sensing measurements allow the quantification of aerosol optical properties (AOPs) at increased spatio-temporal resolutions, they still have limitations retrieving sufficiently accurate AOPs other than the aerosol optical depth (AOD). Surface in situ measurements of AOPs play a crucial role (Hansen et al., 1995) in the reduction of the uncertainty by providing essential information in a more direct way (Andrews et al., 2011; Park et al., 2019).

Continuous, long-term measurements of aerosols, especially in the free troposphere (FT), are needed to understand their long-range transport, trends, and global or regional climate effects (Laj et al., 2009). Aerosols in the FT are spatially more representative than observations within the boundary layer because the lifetime of atmospheric aerosols lifted into the FT can be extended up to several weeks (Kent et al., 1998), and they can travel much faster and further due to the strong prevailing winds (McKendry et al., 2001; Wandinger et al., 2002; Liu et al., 2003; Mattis et al., 2008; Uno et al., 2009).

Measurements of AOPs at the Mauna Loa Observatory (MLO; 19.54°N, 155.58°W, 3397 m above mean sea level) were conducted by the National Oceanic and Atmospheric Administration (NOAA) Earth System Research Laboratory (ESRL) Global Monitoring Division (GMD), as part of the NOAA Federated Aerosol Network (NFAN; Andrews et al., 2019). The MLO has been considered as an ideal location to monitor the FT background aerosol properties because of its geographical location (Lee et al., 1994; Ryan et al., 1997; Perry et al., 1999; Andrews et al., 2019). However, several model simulations and in situ measurements revealed that MLO is affected by both long-range-transported (LRT) aerosols, and aerosols entrained from the local planetary boundary layer (PBL; Mendonca, 1969; Shaw, 1980; Bodhaine et al., 1981; Miller, 1981; Darzi et al., 1982; Merrill et al., 1989; Harris et al., 1990; Bodhaine et al., 1995; Ryan, 1997; Perry et al., 1999; Takekura et al., 2002; Eck et al., 2005; Sharma and Barnes, 2016). To investigate the FT background of aerosol characteristics at MLO, it is necessary to deconvolute the influences from LRT and the local PBL.

In this study, we investigate the aerosol scattering and
absorption properties at MLO from surface in situ measurements. We explore the diurnal variation of AOPs and determine the FT conditions (i.e., by excluding local influences) using Rn-222 concentrations. Seasonal variations and the trend of AOPs are then analyzed according to airmass origin.

METHODS

Hourly mean aerosol scattering coefficient ($\sigma_{sp}$) measured for total suspended particles without size cuts (January 1974–April 2000), and for sub-10 µm particles (April 2000–December 2015) with nephelometers at MLO, were used in this study. This is because the aerosol impactor system, which switches every 6 minutes for measuring sub-10 µm and submicron particles, was installed in April 2000 (Sheridan et al., 2001; Delene and Ogren, 2002). The aerosol absorption coefficient ($\sigma_{ap}$) for sub-10 µm particles measured with filter-based absorption photometers (i.e., particle soot absorption photometer [PSAP] and continuous light absorption photometer [CLAP]) from April 2000 were analyzed. Both $\sigma_{ap}$ and $\sigma_{sp}$ were measured under low relative humidity (RH; < 40%; Sheridan et al., 2001) and were corrected to standard temperature and pressure (STP; i.e., 273.15 K and 1013.25 hPa). All $\sigma_{ap}$ and $\sigma_{sp}$ data (Level 2) were downloaded from NOAA/ESRL/GMD (ftp://ftp.cmdl.noaa.gov/aerosol/mlo/). Detailed descriptions of instruments, data periods, data corrections, and associated uncertainties are listed in Table 1.

Intensive AOPs, such as single-scattering albedo (SSA), scattering Ångström exponent (SÅE), and absorption Ångström exponent (AÅE), were derived from $\sigma_{ap}$ and $\sigma_{sp}$ to examine more detailed aerosol radiative and physical characteristics (Delene and Ogren, 2002). In this study, SSA was calculated at 550 nm (Eq. (1)). Herein, $\sigma_{ap}$ was adjusted to a wavelength ($\lambda$) of 550 nm by using the $1/\lambda$ dependence of aerosol light absorption (van der Hulst, 1957; Bergstrom et al., 2002).

Single Scattering Albedo (550 nm) = 
\[
\frac{\sigma_{sp} (550 \text{ nm})}{\sigma_{sp} (550 \text{ nm}) + \sigma_{ap} (550 \text{ nm})}
\]  

(1)

SÅE was calculated from $\sigma_{ap}$ at 450 and 700 nm wavelengths:

\[
\text{Scattering Ångström Exponent} = \frac{\ln \left( \frac{\sigma_{ap} (450 \text{ nm})}{\sigma_{ap} (700 \text{ nm})} \right)}{\ln \left( \frac{450 \text{ nm}}{700 \text{ nm}} \right)}
\]  

(2)

These intensive AOPs were calculated only if $\sigma_{ap} \geq 1 \text{ Mm}^{-1}$ and $\sigma_{sp} \geq 0.1 \text{ Mm}^{-1}$ to avoid substantial relative uncertainties which were induced when $\sigma_{ap}$ ($\sigma_{sp}$) was close to the detection limit.

Hourly mean Rn-222 volume concentrations (mBq m$^{-3}$) recorded at MLO since 2003 enabled us to identify the time

### Table 1. Descriptions of aerosol scattering and absorption measurements at the Mauna Loa Observatory (MLO).

| Wavelength (nm) | Period | Aerosol scattering coefficient (µm$^{-1}$) | Particle soot absorption coefficient (µm$^{-1}$) |
|-----------------|--------|------------------------------------------|-----------------------------------------------|
| 450, 550, 700   | 1994–2007 | 450, 550, 700, 850 | 450, 550, 700, 850 |
| 450, 550, 700   | 2008–2015 | 450, 550, 700, 850 | 450, 550, 700, 850 |

**Properties**

- NOAA/ESRL/GMD (ftp://ftp.cmdl.noaa.gov/aerosol/mlo/)
- Detailed descriptions of instruments, data periods, data corrections, and associated uncertainties are listed in Table 1.

**Instruments**

- MRI nephelometer (103)
- MÅ nephelometer (Model 3563)
- TSI nephelometer

**Comments**

- Uncertainty at a 95% confidence interval is approximately 33% after corrections are applied (Sherman et al., 2005).
- Median uncertainty at a 95% confidence interval is approximately 35% after corrections are applied (Bond et al., 1999; Ogren et al., 2017).
of the day when the observatory is least perturbed by local influences. Rn-222 is a naturally occurring radioactive gas with a relatively short half-life (3.82 days; Turekian et al., 1977). Additionally, the main influx of Rn-222 to the atmosphere is attributed to the land surface, which is 2–3 orders higher than the oceanic flux (Schery and Huang, 2004). Rn-222 remains in a gaseous state in the atmosphere and is known to be removed solely by its radioactive decay due to its hydrophobicity and nature as a noble gas (Turekian et al., 1977). Therefore, Rn-222 can be considered as an ideal tracer for identifying terrestrial (soil) influences (Chambers et al., 2011, 2013, 2016).

Airmass backward trajectories (BTs) over 10-day periods (240 h), calculated using the NOAA Air Resources Laboratory’s Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model (v4.0; Draxler and Hess, 1998) from the location of MLO, were used to identify the source regions of aerosols at the MLO in FT conditions. National Centers for Environmental Prediction (NCEP)/National Center for Atmospheric Research (NCAR) reanalysis data (gbl) was utilized as an input meteorological field (e.g., horizontal and vertical winds; Kalnay et al., 1996). Each BT was constructed every hour and was paired with hourly mean AOPs to identify the relationship between the AOPs and the origin of the airmass.

The trends of $\sigma_{ap}$ and $\sigma_{ap}$ with their significance over the study period were examined with the Theil-Sen slope method and the Mann-Kendall (MK) test. The Theil-Sen method calculates the slope of the possible trend with a nonparametric approach. It uses the median value of the calculated slopes from all possible pairs of data synched with time information. The Mann-Kendall test is a nonparametric statistical test which is well suited to identify small but monotonic trends. These are typically used because of their insensitivity to missing values and outliers in a time series (Mann, 1945; Kendall, 1975; Gilbert, 1987; Collaud Coen et al., 2013).

RESULTS AND DISCUSSION

The median value of $\sigma_{ap}$ at 550 nm based on measurements collected during a 42-year period (1974–2015) was 0.94 Mm$^{-1}$, and the median value $\sigma_{ap}$ at 550 nm during the period of 2000–2015 was 0.13 Mm$^{-1}$. The overall mean values of $\sigma_{ap}$ and $\sigma_{ap}$ at 550 nm were 2.47 Mm$^{-1}$ and 0.28 Mm$^{-1}$, respectively, with relatively large standard deviations (4.99 Mm$^{-1}$ and 0.45 Mm$^{-1}$). This implies that MLO is operating under the pristine conditions most of the time, but it is intermittently affected by highly aerosol-loaded airmasses.

Diurnal Variation of AOPs and Determination of FT Condition

Fig. 1 shows the diurnal variation of in situ AOPs and Rn-222 concentrations at MLO. $\sigma_{ap}$ exhibited a distinct diurnal variation, with increases in the afternoon hours (14–17 LST) and decreases during the morning hours (8–11 LST). High aerosol loadings, as indicated by $\sigma_{ap}$, during the afternoon hours, can be explained by the prevailing thermally induced (anabatic) winds up the flanks of Mauna Loa Mountain. Elevated Rn-222 concentration—which represents how recently the air mass was in contact with the land surface (Chambers et al., 2011, 2013)—is usually associated with the upslope wind which develops along the ridge of the mountain during the afternoon hours, whereas the upslope wind is also responsible for the influx of the PBL aerosols to MLO (Ryan et al., 1997). Meanwhile, no significant diurnal variation was observed in $\sigma_{ap}$. The transport of scattering-dominant maritime aerosols from PBL to MLO by the aforementioned upslope wind is thought to be the reason for the elevated SSA in the afternoon, since MLO is located on the Big Island of Hawaii, where no particular industrial activities are held (DBEDT, 2019).

Interestingly, Rn-222 concentrations typically decreased between approximately 8 and 11 LST, which is a transition period between dominant, thermally driven katabatic (downslope) and anabatic (upslope) winds (Ryan et al., 1997; Chambers et al., 2013). In this study, we designated these hours of the day (8–11 LST) as Least Locally Influenced (LLI) hours to examine AOPs in FT conditions. A summary of the values of $\sigma_{ap}$ and $\sigma_{ap}$ over 24-h and LLI-h periods is listed in Table 2. Both daily mean and median $\sigma_{ap}$ values were approximately 34% and 32% higher than those of the LLI hours, respectively, whereas the $\sigma_{ap}$ values corresponding to the 24-h and LLI-h periods were not significantly different. Compared to the daily mean, the slightly lower SSA (0.87 ± 0.12) and higher SÅE (1.35 ± 1.24) values during the LLI h (see the white dashed boxes in Fig. 1(b)) suggest that the aerosols in FT conditions are more absorbing and slightly larger.

Seasonal Variation of FT Aerosols

Air Mass Origins

Fig. 2 shows the aerosol optical depth (550 nm) from 13-year Moderate Resolution Imaging Spectroradiometer (MODIS) measurements with selected air mass source regions, and the monthly variation of percentages of air mass origins estimated based on BTs constructed over 10-day periods. The largest fraction of airmasses that reached MLO was from the Pacific Ocean (PO; 43.5%). Approximately 24.7% and 13.1% of airmasses originated from Northeast Asia (NE Asia) and Southeast Asia (SE Asia), respectively, the largest emission sources of natural (dust, biomass burning) and anthropogenic aerosols, as indicated by the MODIS-derived AOD. Several studies reported that aerosol properties at MLO were largely affected by Asian outflow (Bodhaine et al., 1981; Bodhaine, 1995; 1996; Perry et al., 1999; Eck et al., 2005). It is noteworthy that the AOPs at MLO were not much influenced by airmasses that originated from other regions (18.7%; Central America, continents in North Pacific Ocean, North America and continents in the Southern Hemisphere).

MLO is more frequently influenced by Asian airmasses (> 50%) from December to April due to the southward shift of the Intertropical Convergence Zone (ITCZ; Henderson-Sellers and Robinson, 1991; Schneider et al., 2014). By contrast, airmasses from Asia substantially decrease during June–September due to weakening FT westerlies along the Hawaiian Islands by the northward shift of the ITCZ. Instead, airmasses from North and Central America increase, even
Fig. 1. Diurnal variation of the aerosol optical properties (AOPs; $\sigma_{sp}$, $\sigma_{ap}$, SÅE and SSA) and $^{222}$Rn concentration at the Mauna Loa Observatory (MLO). (a) Box-and-whisker plot, whereby the whiskers represent the 10th and 90th percentiles, and the horizontal lines in boxes represent the 25th, 50th, and 75th percentiles of the hourly values. The mean values are denoted with red dots. (b) Annual cycle of the diurnal variability of AOPs. Variables are normalized with the maximum hourly average value of the month. The least locally influenced (LLI) period (8–11 LST) is denoted by the white dotted line.

Table 2. Comparison of aerosol scattering ($\sigma_{sp}$) and absorption ($\sigma_{ap}$) coefficients over 24-h and least locally influenced (LLI)-h periods.

|                                | 24 h (0–24 LST)$^a$ | LLI h (8–11 LST) |
|--------------------------------|----------------------|------------------|
| **Aerosol scattering coefficient** (550 nm, Mm$^{-1}$) |                       |                  |
| Mean                           | 2.47                 | 1.85             |
| Standard deviation             | 4.99                 | 3.43             |
| MAD$^b$ from mean              | 2.48                 | 1.87             |
| Median                         | 0.94                 | 0.71             |
| MAD from median                | 2.03                 | 1.51             |
| **Aerosol absorption coefficient** (550 nm, Mm$^{-1}$) |                       |                  |
| Mean                           | 0.28                 | 0.29             |
| Standard deviation             | 0.45                 | 0.42             |
| MAD from mean                  | 0.26                 | 0.26             |
| Median                         | 0.13                 | 0.14             |
| MAD from median                | 0.22                 | 0.22             |

$^a$ Times are in Hawaiian local standard time (LST = Coordinated Universal Time [UTC] – 10 h).

$^b$ MAD: Mean absolute deviation.
though the MLO is typically located above the trade wind inversion (TWI) layer. This is attributed to the weakened TWI due to the increased thermal instability together with an updraft induced from large-scale circulation (Hastenrath, 1991).

Aerosol Optical Properties

Monthly variations of $\sigma_{ap}$, $\sigma_{sp}$, SÅE, and SSA are shown in Fig. 3. The distinct springtime peaks of $\sigma_{ap}$ and $\sigma_{sp}$ are apparent in both the 24-h and LLI-h periods. Both the $\sigma_{ap}$ and $\sigma_{sp}$ values at 550 nm during the spring months (March–May) were $4.52 \pm 3.75$ Mm$^{-1}$ and $0.49 \pm 0.59$ Mm$^{-1}$, respectively. These were almost twice as large as the annual mean ($\sigma_{ap}$: 2.47 ± 4.99 Mm$^{-1}$; $\sigma_{sp}$: 0.28 ± 0.45 Mm$^{-1}$). Similarly, the values of $\sigma_{ap}$ and $\sigma_{sp}$ in FT conditions (i.e., during LLI hours) during the spring exhibited values approximately twice as large ($3.55 \pm 4.79$ Mm$^{-1}$ and $0.49 \pm 0.52$ Mm$^{-1}$, respectively) as their annual mean values ($\sigma_{ap}$: $1.85 \pm 3.43$ Mm$^{-1}$; $\sigma_{sp}$: $0.29 \pm 0.42$ Mm$^{-1}$). Enhanced $\sigma_{ap}$ and $\sigma_{sp}$ values in FT conditions during the spring can be explained by the FT transport of aerosols, particularly from the Asian continent.

SÅE was relatively low in the spring compared with summer and autumn. This can be explained by the relatively coarse dust particles from NE Asia. Monthly mean SSA at 550 nm ranged between 0.83 and 0.88 from October to April, while it remained > 0.9 during the summer. Observations of SSA smaller than 0.8 during the autumn can be attributed to preferential scavenging of light-scattering aerosols by clouds, fog and/or precipitation at low-$\sigma_{ap}$ and -$\sigma_{sp}$ conditions (Andrews et al., 2011). Frequent transport of light-absorbing aerosols from NE and SE Asia is responsible for low SSA values from January to April.

Figs. 4 and 5 show the monthly variations of $\sigma_{ap}$ and $\sigma_{sp}$ in FT conditions according to the airmass origin, and the contributions of each airmass source region on $\sigma_{ap}$ and $\sigma_{sp}$. We note that the contribution of the airmass origin ($C_{\sigma_{ap}}$) to $\sigma_{ap}$ ($\sigma_{sp}$) was normalized for the $i^{th}$ source region and $j^{th}$ month:

$$C_{\sigma_{ap}}, = \sigma_{ap(q)j} \left( \frac{N_{ij}}{N_j} \right)$$

where $N$ is the number of events. Elevated $\sigma_{ap}$ and $\sigma_{sp}$ values were apparent in the spring with prevailing airmass transportation from NE and SE Asia. It should be noted that monthly variations of $\sigma_{ap}$ and $\sigma_{sp}$ do not always coincide with the frequency of airmass source regions. Compared to the spring, more airmasses from NE and SE Asia reached the MLO in the winter. However, Asian airmasses contribute more to elevated $\sigma_{ap}$ ($\sigma_{sp}$) values in the spring. For example, the contributions of transported aerosols over long ranges from NE Asia (i.e., pollution and Asian dust particles) and from SE Asia (i.e., biomass burning aerosols which is listed...
Airmasses in the mid-troposphere tend to move westward from the late spring due to the influences from the Tibetan High (Liu et al., 2003), so the transport of aerosols from SE Asia to MLO is subsequently decreased. Interestingly, airmasses which travel only within the PO during the spring yield higher $\sigma_{op}$ and $\sigma_{sp}$ values compared to other months (Fig. 4). This is because the Asian aerosols, which are extensively distributed over the Pacific, can also reach MLO. More investigations on aerosol loadings and associated aerosol optical and radiative properties over broad Pacific regions during the spring are thus needed (Brock et al., 2019; Katich et al., 2018).

### Systematic Relationships Among AOPs

We investigated the relationships among AOPs for three major contributing source regions (PO, NE Asia, and SE Asia) to explore the aerosol characteristics, such as their types, sources, and processes (Andrews et al., 2011; Sherman et al., 2015; Schmeisser et al., 2017). Statistical comparisons of $\sigma_{op}$ and $\sigma_{sp}$ for three source regions are listed in Table 3. The highest $\sigma_{op}$ and $\sigma_{sp}$ values were apparent in the NE Asian airmass, whereas the PO airmass yielded the lowest values.

As $\sigma_{op}$ increases, $\sigma_{sp}$ also increases in all three regions (Fig. 6(a)). The higher slope between $\sigma_{op}$ and $\sigma_{sp}$ for the airmass from SE Asia suggests that the aerosols from SE Asia have a lower SSA than others, as shown in Fig. 6(b). Similarly, SSA also gradually increases as $\sigma_{sp}$ increases. Selective scavenging of larger scattering aerosols is possible given that the removal of larger particles generally result in low aerosol concentration with higher absorption parts over the total extinction (Sellergren et al., 2003; Andrews et al., 2011). Contrary to the PO and SE Asia, SÅE gradually decreases with increasing $\sigma_{op}$ for the airmass from NE Asia (Fig. 6(c)). This relationship can be explained by the transport of coarse dust particles from arid and desert areas in NE Asia (Lee et al., 2012). An increasing SÅE with increasing $\sigma_{op}$ values for SE Asian airmasses is likely attributed to the fine-mode biomass burning aerosols (Toledano et al., 2007; Andrews et al., 2011; Schmeisser et al., 2017). SÅE and AÅE between NE and SE Asia are similar, but slightly higher SÅE and lower AÅE were observed in PO airmass (not shown).

### Inter-annual Trend of Aerosol Scattering and Absorption Coefficients

The trends of $\sigma_{op}$ and $\sigma_{sp}$ in FT conditions were calculated for three major source regions (NE Asia, SE Asia, and PO). Fig. 7(a) shows the time series of the annual mean $\sigma_{op}$ and $\sigma_{sp}$ values over the study period. A linear trend and its significance calculated with the Theil-Sen and Mann-Kendall methods is presented in Fig. 7(b). The value of $\sigma_{op}$ increased by approximately $+1.89\%$ per year during the period of 1974–2015. The highest increasing trend of $\sigma_{op}$ since 1974 appeared in the PO air mass ($+2.18\%$ year$^{-1}$), followed by NE Asia ($+2.09\%$ year$^{-1}$) and SE Asia ($+1.22\%$ year$^{-1}$). However, the magnitude of the increasing trend adhered to the order of a) NE Asia ($+0.51$ Mm$^{-1}$ decade$^{-1}$), b) PO ($+0.32$ Mm$^{-1}$ decade$^{-1}$), and c) SE Asia ($+0.27$ Mm$^{-1}$ decade$^{-1}$).

---

**Fig. 3.** Monthly variation of AOPs ($\sigma_{op}$, $\sigma_{sp}$, SÅE and SSA) observed at the MLO. The whiskers represent the 10th and 90th percentiles, and the horizontal lines in boxes represent the 25th, 50th, and 75th percentiles of monthly data. Monthly averages are denoted as dots. Percentiles of 24-h measurements are shown in red, while percentiles of LLI-h data are shown in blue.

---

*References*: Brock et al., 2019; Katich et al., 2018; Liu et al., 2003; Lee et al., 2017; Nam et al., 2018; Park et al., 2019.
Fig. 4. Monthly variations of $\sigma_{sp}$ and $\sigma_{ap}$ for the air masses from the PO, NE Asia, and SE Asia regions. Cross lines in boxes represent the 25th, 50th, and 75th percentiles, and whiskers represent the 10th and 90th percentiles. Mean values of each month are denoted by asterisks. Data for $\sigma_{sp}$ and $\sigma_{ap}$ from the period of 2000–2015 are utilized.

Fig. 5. Monthly variation of source region contributions on $\sigma_{sp}$ and $\sigma_{ap}$ at the MLO. PO, NE Asia, SE Asia, and other regions (OR) are denoted with blue, red, green, and gray colors, respectively.
Table 3. Comparison of aerosol scattering and absorption coefficients (550 nm) among different source regions.

| Properties                     | Pacific Ocean | Northeast Asia | Southeast Asia | Other regions^a |
|--------------------------------|---------------|----------------|----------------|-----------------|
| Aerosol scattering coefficient | Mean          | 1.457          | 2.456          | 2.243           | 1.828           |
|                                | Median        | 0.64           | 0.93           | 0.83            | 0.68            |
| (550 nm, Mm^-1)                | Standard error| 0.034          | 0.064          | 0.082           | 0.060           |
| Aerosol absorption coefficient | Mean          | 0.229          | 0.358          | 0.352           | 0.273           |
| (550 nm, Mm^-1)                | Median        | 0.126          | 0.187          | 0.161           | 0.012           |
|                                | Standard error| 0.0078         | 0.0133         | 0.0171          | 0.015           |

^a Includes North America, Central America, Southern Hemispheric continents and North Pacific Continents.

Fig. 6. Systematic relationship between the FT AOPs at the MLO analyzed according to the source region. Red, green, and blue colors respectively indicate the NE Asia, SE Asia, and PO regions. (a–c) Systematic relations among σ_sp and other AOPs, such as σ_ap, single-scattering albedo (SSA), and scattering Ångström exponent (SÅE). The average values of variables correspond to each σ_sp bin, which is divided in 2 Mm^-1 intervals, are denoted by filled diamonds with the respective colors used for each source region. Horizontal lines in boxes represent standard errors. Bins with more than 20 valid measurements are analyzed. Linear regression lines over each source region are denoted with dotted lines using the respective colors for the studied regions.

Fig. 7. (a) Time series of annual average σ_sp and σ_ap values according to the source region. Each source region is depicted in blue (PO), red (NE Asia), and green (SE Asia) colors. (b) Trends of σ_sp and σ_ap values according to the air mass origins. Trends and their statistical significances are calculated with the Theil-Sen slope method and the Mann-Kendall test, respectively. Significant trends at 99% (95%) confidence levels are denoted as circles (triangles), while crosses denote insignificant trends at a 95% confidence level.
These first two trends (a, b) are significant at a 99% confidence level, and the last trend (c) at a 95% confidence. Both the $\sigma_p$ and $\sigma_a$ values yield positive trends in all three regions during the period of 2000–2015, but both are insignificant at a 95% confidence level. Overall, $\sigma_p$ and $\sigma_a$ at MLO is experiencing greater influence from LRT aerosol plumes. Especially, aerosol transport from NE Asia, associated with increasing anthropogenic emission due to the economic growth (and with some natural variation) was the most prominent contributing source for the increasing trend of the extensive AOPs (Liu et al., 2003; Guo et al., 2011; Kim et al., 2011; Chen and Wang, 2015). The positive trend in PO airmass is attributable to enhanced outflow of pollution aerosols from NE Asia, which finally reaches MLO. This is supported by the concurrent peak of $\sigma_p$ and $\sigma_a$ found during boreal spring when the source of air mass is PO, while the only possible FT source of aerosols are FT transport originating from NE and SE Asia.

**CONCLUSIONS**

We investigated diurnal, monthly, and inter-annual variations in the aerosol scattering coefficient ($\sigma_p$: 1974–2015) and the absorption coefficient ($\sigma_a$: 2000–2015) at the MLO using surface in situ measurements. The major findings of this study are summarized below:

- The value of $\sigma_p$ decreased during the hours of 8–11 LST (1.85 ± 3.43 Mm⁻¹ at 550 nm) but increased during the afternoon (3.72 ± 7.63 Mm⁻¹ at 550 nm; 14–17 LST) due to the development of upslope boundary layer winds. No distinct diurnal variation was observed in $\sigma_a$.

- The highest $\sigma_p$ and $\sigma_a$ values appeared when the air masses originated in NE Asia ($\sigma_p$: 2.46 Mm⁻¹; $\sigma_a$: 0.36 Mm⁻¹), followed by SE Asia ($\sigma_p$: 2.24 Mm⁻¹; $\sigma_a$: 0.35 Mm⁻¹) and the PO ($\sigma_p$: 1.46 Mm⁻¹; $\sigma_a$: 0.23 Mm⁻¹).

- NE Asia and SE Asia were the most prominent sources of air masses during the winter, but their contributions to $\sigma_p$ and $\sigma_a$ values peaked during the spring.

- A distinct increase in the values of $\sigma_p$ and $\sigma_a$ during the spring under FT conditions (8–11 LST) was attributed to long-range-transported dust and pollution aerosols from NE Asia and biomass burning aerosols from SE Asia.

- The largest increasing trend for $\sigma_a$ after 1974 was attributed to air masses from the PO (+2.18% year⁻¹), followed by NE Asia (+2.09% year⁻¹) and SE Asia (+1.22% year⁻¹). However, the increasing trend’s magnitude adhered to the order of a) NE Asia (+0.51 Mm⁻¹ decade⁻¹), b) the PO (+0.32 Mm⁻¹ decade⁻¹), and c) SE Asia (+0.27 Mm⁻¹ decade⁻¹). Both the $\sigma_p$ and $\sigma_a$ values showed positive trends for all three regions over the period of 2000–2015, but these values were insignificant at a 95% confidence level.

Long-term, continuous climate-relevant aerosol measurements at the MLO are needed in the future to better estimate the direct aerosol radiative effects related to emissions from Asia. In particular, simultaneous measurements of aerosol chemical components will be very helpful in identifying the aerosol sources.

**ACKNOWLEDGEMENTS**

This study was supported by the Basic Science Research Program through the National Research Foundation of Korea (NRF) funded by the Ministry of Education (2017R1D1A1B06032548) and the Korea Meteorological Administration Research and Development Program under Grant KMI2018-01111. Authors are also thankful to operators and technicians at Mauna Loa Observatory for supporting the measurements by conducting maintenance and calibrations of instruments.

**SUPPLEMENTARY MATERIAL**

Supplementary data associated with this article can be found in the online version at http://www.aaqr.org.

**REFERENCES**

Anderson, T.L. and Ogren, J.A. (1998). Determining aerosol radiative properties using the TSI 3563 integrating nephelometer. *Aerosol Sci. Technol.* 29: 57–69. https://doi.org/10.1080/02786829808965551

Andrews, E., Ogren, J.A., Bonasoni, P., Marinoni, A., Cuevas, E., Rodríguez, S., Sun, J.Y., Jaffe, D.A., Fischer, E.V., Baltensperger, U., Weingartner, E., Coen, M.C., Sharma, S., Macdonald, A.M., Leaitch, W.R., Lin, N.H., Laj, P., Arsov, T., Kalapov, I., Jefferson, A. and Sheridan, P. (2011). Climatology of aerosol radiative properties in the free troposphere. *Atmos. Res.* 102: 365–393. https://doi.org/10.1016/j.atmosres.2011.08.017

Andrews, E., Sheridan, P.J., Ogren, J.A., Hageman, D., Jefferson, A., Wendell, J., Alastuey, A., Alados-Arboledas, L., Bergin, M., Ealo, M., Gannet Hallar, A., Hoffer, A., Kalapov, I., Keywood, M., Kim, J, Kim, S.W., Kolonjari, F., Labuschagne, C., Lin, N.H., Macdonald, A., Mayol-Bracero, O.L., McCubbin, I.B., Pandolfi, M., Reisen, F., Sharma, S., Sherman, J.P., Sorríbas, M. and Sun, J. (2019). Overview of the NOAA/ESRL federated aerosol network. *Bull. Am. Meteorol. Soc.* 100: 123–135. https://doi.org/10.1175/BAMS-D-17-0175.1

Bergstrom, R.W., Russell, P.B. and Hignett, P. (2002). Wavelength dependence of the absorption of black carbon particles: Predictions and results from the TARFOX experiment and implications for the aerosol single scattering albedo. *J. Atmos. Sci.* 59: 567–577. https://doi.org/10.1175/1520-0469(2002)059<0567:WDATAO>2.0.CO;2

Bey, I., Jacob, D.J., Logan, J.A. and Yantosca, R.M. (2001). Asian chemical outflow to the Pacific in spring: Origins, pathways, and budgets. *J. Geophys. Res.* 106: 23097–23131. https://doi.org/10.1029/2001JD000806

Bodhaine, B.A., Mendonça, B.G., Harris, J.M. and Miller, M. (1981). Seasonal variations in aerosols and atmospheric transmission at Mauna Loa Observatory. *J. Geophys. Res.* 86: 7395–7398. https://doi.org/10.1029/JC086iC08p07395

Bodhaine, B.A. (1995). Aerosol absorption measurements at Barrow, Mauna Loa and aerosol have also been measured continuously for many years is consistent with
specific absorption of BC on the aethalometer specific absorption atmosphere measurements of the wavelength dependence. J. Geophys. Res. 100: 8967–8975. https://doi.org/10.1029/95JD00513

Bodhaine, B.A. (1996). Aerosol measurements during the Mauna Loa Photochemistry Experiment 2. J. Geophys. Res. 101: 14757–14765. https://doi.org/10.1029/95JD02045

Bond, T.C., Anderson, T.L. and Campbell, D. (1999). Calibration and intercomparison of filter-based measurements of visible light absorption by aerosols. Aerosol Sci. Technol. 30: 582–600. https://doi.org/10.1080/027868299304435

Boucher, O., Randall, D., Artaxo, P., Bretherton, C., Feingold, G., Forster, P., Kerminen, V.M., Kondo, Y., Liao, H., Lohmann, U., Rasch, P., Satheesh, S.K., Sherwood, S., Stevens, B. and Zhang, X.Y. (2013). Clouds and Aerosols. In Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change, Stocker, T.F., Qin, D., Plattner, G.K., Tignor, M., Allen, S.K., Boschung, J., Nauels, A., Xia, Y., Bex, V. and Midgley, P.M. (Eds.), Cambridge University Press.

Brock, C.A., Williamson, C., Kucp, A., Froyd, K.D., Erdesz, F., Wagner, N., Richardson, M., Schwarz, J.P., Gao, R.S., Katich, J.M., Campuzano-Jost, P., Nault, B.A., Schroder, J.C., Jimenez, J.L., Weinzierl, B., Dollner, M., Bui, T. and Murphy, D.M. (2019). Aerosol size distributions during the Atmospheric Tomography Mission (ATom): Methods, uncertainties, and data products. Atmos. Meas. Tech. 12: 3081–3099. https://doi.org/10.5194/amt-12-3081-2019

Cai, W., Li, K., Liao, H., Wang, H. and Wu, L. (2017). Weather conditions conducive to Beijing severe haze more frequent under climate change. Nat. Clim. Change 7: 257–262. https://doi.org/10.1038/nclimiate3249

Chambers, S., Williams, A.G., Zahorowski, W., Griffiths, A. and Crawford, J. (2011). Separating remote fetch and local mixing influences on vertical radon measurements in the lower atmosphere. Tellus B 63: 843–859. https://doi.org/10.1111/j.1600-0889.2011.00565.x

Chambers, S.D., Zahorowski, W., Williams, A.G., Crawford, J. and Griffiths, A.D. (2013). Identifying tropospheric baseline air masses at mauna loa observatory between 2004 and 2010 using radon-222 and back trajectories. J. Geophys. Res. 118: 992–1004. https://doi.org/10.1029/2012JD018212

Chambers, S.D., Williams, A.G., Conen, F., Griffiths, A.D., Reimann, S., Steinbacher, M., Krummel, P.B., Steele, L.P., van der Schoot, M.V., Galbally, I.E., Molloy, S.K., Barnes, J.E. (2016). Towards a universal “Baseline” characterisation of air masses for high- and low-altitude observing stations using radon-222. Aerosol Air Qual. Res. 16: 885–899. https://doi.org/10.4209/aaq.2015.06.0391

Chen, H. and Wang, H. (2015). Haze Days in North China and the associated atmospheric circulations based on daily visibility data from 1960 to 2012. J. Geophys. Res. 120: 5895–5909. https://doi.org/10.1002/2015JD023225

Collaud Coen, M., Andrews, E., Asmi, A., Baltensperger, U., Bukowiecki, N., Day, D., Fiebig, M., Fjaeraa, A.M., Flentje, H., Hyvärinen, A., Jefferson, A., Jennings, S.G., Kouvarakis, G., Lihavainen, H., Lund Myhre, C., Malm, W.C., Mihapopoulos, N., Molenaar, J.V., O’Dowd, C., Ogren, J.A., Schichtel, B.A., Sheridan, P., Virkkula, A., Weingartner, E., Weller, R. and Laj, P. (2013). Aerosol decadal trends-Part 1: In-situ optical measurements at GAW and IMPROVE stations. Atmos. Chem. Phys. 13: 869–894. https://doi.org/10.5194/acp-13-869-2013

Darzi, M. and Winchester, W. (1982). Aerosol characteristics at Mauna Loa Observatory, Hawaii, after east Asian dust storm episodes. J. Geophys. Res. 87: 1251–1258. https://doi.org/10.1029/JC087iC02p01251

Delene, D.J. and Ogren, J.A. (2002). Variability of aerosol optical properties at four North American surface monitoring sites. J. Atmos. Sci. 59: 1135–1150. https://doi.org/10.1175/1520-0469(2002)059<1135:VOAOPA>2.0.CO;2

Draxler, R.R. and Hess, G.D. (1998). An overview of the HYSPLIT_4 modelling system for trajectories, dispersion, and deposition. Aust. Meteorol. Mag. 47: 295–308.

Eck, T.F., Holben, B.N., Dubovik, O., Smirnov, A., Goloub, P., Chen, H.B., Chatenet, B., Gomes, L., Zhang, X.Y., Tsay, S.C., Ji, Q., Giles, D. and Slutsker, I. (2005). Columnar aerosol optical properties at AERONET sites in central eastern Asia and aerosol transport to the tropical mid-Pacific. J. Geophys. Res. 110: 1–18. https://doi.org/10.1029/2004JD005274

Garreaud, R.D. (2001). Subtropical cold surges: Regional aspects and global distribution. Int. J. Climatol. 21: 1181–1197. https://doi.org/10.1002/joc.687

Gilbert, R.O. (1987). Statistical Methods for Environmental Pollution Monitoring. Van Nostrand Reinhold Company Inc., New York.

Guo, J.P., Zhang, X.Y., Wu, Y.R., Zhaxi, Y., Che, H.Z., La, B., Wang, W. and Li, X.W. (2011). Spatio-temporal variation trends of satellite-based aerosol optical depth in China during 1980-2008. Atmos. Environ. 45: 6802–6811. https://doi.org/10.1016/j.atmosenv.2011.03.068

Hansen, J., Rossw, W., Carlson, B., Lacin, A., Travis, L., Del Genio, A., Fung, I., Cairns, B., Mishchenko, M. and Sato, M. (1995). Forcing and Feedbacks. Clim. Change. 31: 247–271.

Harris, J.M. and Kahl, J.D. (1990). A descriptive atmospheric transport climatology for the Mauna Loa Observatory, using clustered trajectories. J. Geophys. Res. 95: 13651–13667. https://doi.org/10.1029/JD095iD09p13651

Hastenrath, S. (1991). Climate Dynamics of the Tropics. Springer. Netherlands. https://doi.org/10.1007/978-94-011-3156-8

Haywood, J.M. and Ramaswamy, V. (1998). Global sensitivity studies of the direct radiative forcing due to anthropogenic sulfate and black carbon aerosols. J. Geophys. Res. 103: 6043–6058. https://doi.org/10.1029/ 97JD03426

Henderson-Sellers, A. and Robinson, P.J. (1991). Contemporary climatology. John Wiley. New York.

Jacobson, M.Z. (2002). Control of fossil-fuel particulate black carbon and organic matter, possibly the most
effective method of slowing global warming. J. Geophys. Res. 107: ACH 16-1-ACH 16-22. https://doi.org/10.1029/2001JD001376

Kalnay, E., Kanamitsu, M., Kistler, R., Collins, W., Deaven, D., Gandin, L., Iredell, M., Saha, S., White, G., Woollen, J., Zhu, Y., Chelliah, M., Ebisuzaki, W., Higgins, W., Janowiak, J., Mo, K.C., Ropelewski, C., Wang, J., Leetmaa, A., Reynolds, R., Jenne, R. and Joseph, D. (1996). The NCEP NCAR 40-Year Reanalysis Project. Bull. Am. Meteorol. Soc. 77: 437–472. https://doi.org/10.1175/1520-0477(1996)077<0437:TYNRCP>2.0.CO;2

Katich, J.M., Samset, B.H., Paul Bui, T., Dollner, M., Froyd, K.D., Campuzano-Jost, P., Nault, B.A., Schroder, J.C., Weinzierl, B. and Schwarz, J.P. (2018). Strong contrast in remote black carbon aerosol loadings between the Atlantic and Pacific basins. J. Geophys. Res. 123: 13386-13395. https://doi.org/10.1002/2018JD029206

Kendall, M.G. (1975). Rank correlation methods. 4th ed. Charles Griffin. London.

Kent, G.S., Trepte, C.R. and Lucker, P.L. (1998). Long-term stratospheric aerosol and gas experiment I and II measurements of upper tropospheric aerosol extinction. J. Geophys. Res. 103: 28863–28874. https://doi.org/10.1029/98JD01395

Kim, N.K., Kim, Y.P. and Kang, C.H. (2011). Long-term trend of aerosol composition and direct radiative forcing due to aerosols over Gosaon: TSP, PM$_{10}$, and PM$_{2.5}$ dust between 1992 and 2008. Atmos. Environ. 45: 6107–6115. https://doi.org/10.1016/j.atmosenv.2011.08.051

Laj, P., Klausen, J., Bilde, M., Plaß-Duelmer, C., Pappalardo, G., Clerbaux, C., Baltensperger, U., Hjorth, J., Simpson, D., Reimann, S., Coheur, P.F., Richter, A., De Mazière, M., Rudich, Y., McFiggans, G., Torseth, K., Wiedensohler, A., Morin, S., Schulz, M., … Zardini, A.A. (2009). Measuring atmospheric composition change. Atmos. Environ. 43: 5351–5414. https://doi.org/10.1016/j.atmosenv.2009.08.020

Lee, G., Merrill, J.T. and Huebert, J. (1994). Variation of free tropospheric total nitrate at Mauna Loa Observatory, Hawaii. J. Geophys. Res. 99: 12821-12831. https://doi.org/10.1029/94JD00146

Lee, H.H., Bar-Or, R.Z. and Wang, C. (2017). Biomass burning aerosols and the low-visibility events in Southeast Asia. Atmos. Chem. Phys. 17: 965–980. https://doi.org/10.5194/acp-17-965-2017

Lee, S., Yoon, S.C., Kim, S.W., Kim, Y.P., Ghim, Y.S., Kim, J.H., Kang, C.H., Kim, Y.J., Chang, L.S. and Lee, S.J. (2012). Spectral dependency of light scattering/absorption and hygroscopicity of pollution and dust aerosols in Northeast Asia. Atmos. Environ. 50: 246–254. https://doi.org/10.1016/j.atmosenv.2011.12.026

Liu, H., Jacob, D.J., Bey, I., Yantosca, R.M., Duncan, B.N. and Sachse, G.W. (2003). Transport pathways for Asian pollution outflow over the Pacific: Interannual and seasonal variations. J. Geophys. Res. 108: GTE 7-1–GTE 7-15. https://doi.org/10.1029/2002JD003102

Mann, H.B. (1945). Nonparametric tests against trend. Econometrica 13: 245–259.

Mattis, I., Müller, D., Ansmann, A., Wandinger, U., Preißler, J., Seifert, P. and Tesche, M. (2008). Ten years of multiwavelength Raman lidar observations of free-tropospheric aerosol layers over central Europe: Geometrical properties and annual cycle. J. Geophys. Res. 113: D20202. https://doi.org/10.1029/2007JD009636

McKendry, I.G., Hacker, J.P., Stull, R., Sakiyama, S., Mignacca, D. and Reid, K. (2001). Long-range transport of Asian dust to the Lower Fraser Valley, British Columbia, Canada. J. Geophys. Res. 106: 18361–18370. https://doi.org/10.1029/2000JD900359

Mendoza, B.G. (1969). Local wind circulation on the slope of Mauna Loa. J. Appl. Meteorol. 8: 533–541. https://doi.org/10.1175/1520-0450(1969)008<0533:LWCO>2.0.CO;2

Merrill, J.T., Uematsu, M. and Bleck, R. (1989). Meteorological analysis of long range transport of mineral aerosols over the North Pacific. J. Geophys. Res. 94: 8584–8598. https://doi.org/10.1029/JD0941D60p08584

Miller, J.M. (1981). A five-year climatology of back trajectories from the Mauna Loa Observatory, Hawaii. Atmos. Environ. 15: 1553–1558. https://doi.org/10.1016/0004-6981(81)90138-4

Nam, J., Kim, S.W., Park, R.J., Park, J.S. and Park, S.S. (2018). Changes in column aerosol optical depth and ground-level particulate matter concentration over East Asia. Air Qual. Atmos. Health 11: 49–60. https://doi.org/10.1007/s11869-017-0517-5

Ogren, J.A. (2010). Comment on “Calibration and intercomparison of filter-based measurements of visible light absorption by aerosols” Aerosol Sci. Technol. 44: 589–591. https://doi.org/10.1080/02786826.2010.482111

Ogren, J.A., Wendell, J., Andrews, E. and Sheridan, P.J. (2017). Continuous light absorption photometer for long-term studies. Atmos. Meas. Tech. 10: 4805–4818. https://doi.org/10.5194/amt-10-4805-2017

Park, S., Kim, S.W., Lin, N.H., Pani, S.K., Sheridan, P.J. and Andrews, E. (2019). Variability of aerosol optical properties observed at a polluted marine (Gosan, Korea) and a high-altitude mountain (Lulin, Taiwan) site in the Asian continental outflow. Aerosol Air Qual. Res. 19: 1272–1283. https://doi.org/10.4209/aaqr.2018.11.0416

Perry, K.D., Cahill, T.A., Schnell, R.C. and Harris, J.M. (1999). Long-range transport of anthropogenic aerosols to the National Oceanic and Atmospheric Administration baseline station at Mauna Loa Observatory, Hawaii. J. Geophys. Res. 104: 18521–18533. https://doi.org/10.1029/1998JD000883

Research and Economic Analysis Division of the State Department of Business, Economic Development & Tourism (DBEDT) (2019). Quarterly statistical and economic report-State of Hawaii. 3rd Quarter 2019. Honolulu.

Ryan, S. (1997). The wind field around Mauna Loa derived from surface and balloon observations. J. Geophys. Res. 102: 10711–10725. https://doi.org/10.1029/97JD00646

Schery, S.D. and Huang, S. (2004). An estimate of the global distribution of radon emissions from the ocean. Geophys. Res. Lett. 31: L19104. https://doi.org/10.1029/2004GL021051
Schmeisser, L., Andrews, E., Ogren, J.A., Sheridan, P., Jefferson, A., Sharma, S., Kim, J.E., Sherman, J.P., Sorribas, M., Kalapov, I., Arsov, T., Angelov, C., Mayol-Bracero, O.L., Labuschagne, C., Kim, S.W., Hoffer, A., Lin, N.H., Chia, H.P., Bergin, M., Sun, J., Liu, P. and Wu, H. (2017). Classifying aerosol type using in-situ surface spectral aerosol optical properties. Atmos. Chem. Phys. 17: 12097–12120. https://doi.org/10.5194/acp-17-12097-2017

Schneider, T., Bischoff, T. and Haug, G.H. (2014). Migrations and dynamics of the intertropical convergence zone. Nature 513: 45–53. https://doi.org/10.1038/nature13636

Sellegri, K., Laj, P., Dupuy, R., Legrand, M., Preunkert, S. and Putaud, J.P. (2003). Size-dependent scavenging efficiencies of multicomponent atmospheric aerosols in clouds. J. Geophys. Res. 108: AAC 3-1–AAC 3-15. https://doi.org/10.1029/2002JD002749

Sherman, J.P., Sheridan, P.J., Ogren, J.A., Andrews, E., Hageman, D., Schmeisser, L., Jefferson, A. and Sharma, S. (2015). A multi-year study of lower tropospheric aerosol variability and systematic relationships from four North American regions. Atmos. Chem. Phys. 15: 12487–12517. https://doi.org/10.5194/acp-15-12487-2015

Uno, I., Nakajima, T., Higurashi, A. and Sano, I. (2002). Modeling study of long-range transport of Asian dust and anthropogenic aerosols from East Asia. Geophys. Res. Lett. 29: 11-1-11-4. https://doi.org/10.1029/2002GL016251

Toledano, C., Cachorro, V.E., Berjon, A., de Frutos, A.M., Sorribas, M., de la Morena, B.A. and Goloub, P. (2007). Aerosol optical depth and Ångström exponent climatology at El Arenosillo AERONET site (Huelva, Spain). Q. J. R. Meteorolog. Soc. 133: 795–807. https://doi.org/10.1002/qj.54

Received for review, November 18, 2019
Revised, February 24, 2020
Accepted, April 3, 2020