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Investigation of Aerosol Peak Height Effect on PBL and Volcanic Air Mass Factors for SO₂ Column Retrieval from Space-Borne Hyperspectral UV Sensors

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Abstract: We investigate the effects of aerosol peak height (APH) and various parameters on the air mass factor (AMF) for SO₂ retrieval. Increasing aerosol optical depth (AOD) leads to multiple scattering within the planetary boundary layer (PBL) and an increase in PBL SO₂ AMF. However, under high AOD conditions, aerosol shielding effects dominate, which causes the PBL SO₂ AMF to decrease with increasing AOD. The height of the SO₂ layer and the APH are found to significantly influence the PBL SO₂ AMF under high AOD conditions. When the SO₂ and aerosol layers are of the same height, aerosol multiple scattering occurs dominantly within the PBL, which leads to an increase in the PBL SO₂ AMF. When the APH is greater than the SO₂ layer height, aerosol shielding effects dominate, which decreases the PBL SO₂ AMF. When the SO₂ and aerosol layers are of the same height under low AOD and solar zenith angle (SZA) conditions, increased surface reflectance is found to significantly increase the PBL SO₂ AMF. However, high AOD dominates the surface reflectance contribution to PBL SO₂ AMF. Under high SZA conditions, Rayleigh scattering contributes to a reduction in the light path length and PBL SO₂ AMF. For volcanic SO₂ AMF, high SZA enhances the light path length within the volcanic SO₂ layer, as well as the volcanic SO₂ AMF, because of the negligible photon loss by Rayleigh scattering at high altitudes. High aerosol loading and an APH that is greater than the SO₂ peak height lead to aerosol shielding effects, which reduce the volcanic SO₂ AMF. The SO₂ AMF errors are also quantified as a function of uncertainty in the input data of AOD, APH, and surface reflectance. The SO₂ AMF sensitivities and error analysis provided here can be used to develop effective error reduction strategies for satellite-based SO₂ retrievals.

Keywords: SO₂ AMF; air mass factor; aerosol height; aerosol layer; trace gas; remote sensing; satellite measurement

1. Introduction

Through the formation of gas-phase sulfuric acid and sulfate aerosol, sulfur dioxide (SO₂) plays an important role in atmospheric chemistry on a global scale and affects short-term pollution as well as climate forcing [1,2]. Natural emissions of volcanic and biogenic dimethyl sulfide account for only ~30% of total global SO₂ emissions, whereas >70% is derived from anthropogenic sources including

fossil fuel combustion and metal smelting [3,4]. The SO₂ emitted from anthropogenic sources is likely to be present in the planetary boundary layer (PBL), particularly near the surface. To date, SO₂ in the PBL has been regionally or globally monitored by multiple hyperspectral ultraviolet (UV) satellite sensors including the Global Ozone Monitoring Experiment (GOME) [5,6], the Scanning Imaging Absorption Spectrometer for Atmospheric Cartography (SCIAMACHY) [7,8], GOME-2 [9–11], the Ozone Monitoring Instrument (OMI) [12,13], the Ozone Mapping and Profiler Suite (OMPS) [14,15], and the TROPOspheric Monitoring Instrument (TROPOMI) [16]. The differential optical absorption spectroscopy (DOAS) spectral fitting method has been widely used to retrieve SO₂ column amounts from radiance data measured by these hyperspectral satellite sensors [2,5–7,9–11,17–23].

Spectral fitting methods such as the DOAS or Principal Component Analysis (PCA) techniques are first used to retrieve the SO₂ slant column density (SCD), which is the integral part of the SO₂ concentration that is present over the path between a light source and the sensor [24]. SO₂ vertical column density (VCD) is obtained by dividing the SO₂ SCD into the SO₂ air mass factor (AMF). When the SO₂ AMF is simulated using a radiative transfer model (RTM), multiple input parameters are required to calculate the SO₂ AMF. Uncertainties in the input data used for these parameters can lead to errors in the AMF calculations. Several studies have investigated the dependence of volcanic SO₂ AMF on the parameters used in its calculation. Thomas et al. [23] reported the dependence of volcanic SO₂ AMF on aerosol optical depth (AOD), aerosol type, and SO₂ plume height. The effects of solar zenith angle (SZA), SO₂ profile, and albedo on the volcanic SO₂ AMF were studied by Khokhar et al. [6]. Richter et al. [10] investigated the dependence of SO₂ AMF on wavelength, SZA, O₃ VCD, SO₂ VCD, and surface reflectivity for a layer with elevated volcanic SO₂. Theys et al. [25] reported the SO₂ vertical distribution effect on volcanic and PBL SO₂ AMF. Finally, Lee et al. [26] carried out a PBL SO₂ AMF error analysis for surface reflectance, aerosol properties, cloud fraction, and SO₂ vertical distribution.

These previous studies have contributed to a general quantitative understanding of the effects of various parameters on SO₂ AMF. However, although aerosol scattering and absorption have important effects on AMF, especially over the short UV wavelength interval of 300–325 nm where PBL SO₂ AMF is usually calculated, the cumulative effects of aerosol vertical location and AOD along with other parameters (e.g., surface reflectance and the ozone column density) on SO₂ AMF have not been studied. In addition, few studies have investigated the influence of the parameters on PBL SO₂ AMF and the effects of their associated uncertainties on PBL SO₂ retrieval error, despite the fact that accurate PBL SO₂ retrievals are required over regions where coal-related industrial activities are prevalent. Quantitative information for each parameter's contribution to SO₂ AMF uncertainty will be useful for reducing errors in SO₂ retrievals. The aim of this study is to investigate the simultaneous effects of aerosol peak height (APH), AOD, geometric information, SO₂ vertical profile (PBL vs. volcanic), ozone amount, ozone vertical profile, and surface reflectance on both PBL and volcanic SO₂ AMF. This study also quantifies SO₂ AMF errors caused by input data uncertainty for APH and other parameters.

2. Methods

The AMF is applied to convert SCD into vertical column density (VCD; $AMF = SCD/VCD$). The AMF can be calculated using the formulation of Palmer et al. [27], and can be expressed in terms of the scattering weight and shape factor, as follows:

$$AMF = AMF_G \int_0^{\infty} \omega'(z) S'_z(z) dz \quad (1)$$

where AMF_G is the geometric AMF, $\omega'(z)$ is the scattering weight, and $S'_z(z)$ is the shape factor for each layer. The scattering weight is the sensitivity of the backscattered spectrum to the abundance of the absorber at each layer, and the shape factor is a normalized vertical profile of number density [27]. The scattering weight is affected by trace gas absorption optical depth, scattering due to air molecules (Rayleigh), and the aerosol extinction optical depth (Mie scattering) [28]. The SO₂ AMF depends on the SO₂ vertical profile, surface albedo, observational geometry (solar zenith, viewing zenith, and relative

azimuth angles), total ozone column, aerosols, and clouds [6,10,23,25,26]. Here, the SO₂ AMF was calculated using the linearized pseudo-spherical scalar and vector discrete ordinate radiative transfer model (VLIDORT, version 2.6). A detailed description of AMF computation using VLIDORT can be found in Spurr and Christi [28]. The PBL SO₂ AMF was computed for 318 nm, which is representative of the wavelength interval of 310–326 nm and includes the strong SO₂ absorption band that peaks between about 310 and 313 nm, and which was used for the spectral fitting. Although SO₂ retrievals in this fitting window are suitable for small anthropogenic SO₂ column amounts, the retrieval accuracy for large volcanic SO₂ amounts is limited by saturation of the SO₂ SCD retrievals caused by strong SO₂ absorption [2,10,22,29]. Therefore, to calculate the volcanic SO₂ AMF, we used a wavelength of 375 nm for use with the fitting window of 360–390 nm employed to retrieve SO₂ columns caused by volcanic eruption [2,18,22].

Figure 1 summarizes the SO₂ AMF calculation. Scattering weight was computed using the AOD, single scattering albedo (SSA), APH, half width (HW) of aerosol vertical distribution, surface reflectance, viewing zenith angle (VZA), and SZA. We calculated the shape factor using the SO₂ vertical profile and its absorption cross-section at 298 K [30].

Figure 1. Flow chart of SO₂ air mass factor (AMF) calculation.

2.1. Vertical Distributions of SO₂ and O₃

The O₃ vertical profiles used for the SO₂ AMF calculations are shown in Figure 2. The nine O₃ vertical distributions obtained from the TOMS-V8 O₃ profile climatology, which vary with latitude and O₃ column, are used to verify the effect of the O₃ vertical profile and vertical column densities (VCD) on the SO₂ scattering weights. A detailed description of the TOMS O₃ climatology can be found in Bhartia and Wellemeyer [31], McPeters et al. [32], and Wellemeyer et al. [33]. As the general SO₂ vertical profile is unknown [2,22], a hypothetical SO₂ profile is used to determine the SO₂ shape factor

for anthropogenic pollution (Figure 3a). For anthropogenic SO_2 , SO_2 box concentration profiles of 1 km thickness are arranged from the surface to a height of 1 km in the PBL. For the volcanic SO_2 case, we chose Iceland's Eyjafjallajökull (63.63°N , 19.62°W ; 1666-m altitude) eruption on 14 April 2010. This eruption ejected a SO_2 plume 8–13 km high into the atmosphere [11,34]. Therefore, for the volcanic SO_2 case, it was assumed that a total SO_2 column of 200 DU was distributed throughout a 1 km box profile from 9 to 10 km in the lower stratosphere (Figure 3b).

Figure 2. O_3 vertical distributions used for the SO_2 AMF calculation.

Figure 3. SO_2 and aerosol vertical distributions used for the (a) planetary boundary layer (PBL) and (b) volcanic SO_2 AMF calculations. The half width is 3 km, and the lower and upper limits for the aerosol height are set to surface level and 10 km, respectively.

2.2. Aerosols

AOD, aerosol upper limit, aerosol lower limit, and APH are used to determine the aerosol vertical profiles. To quantify the effects of aerosols on SO_2 AMF, we used the AOD, APH, aerosol upper limit, and aerosol lower limit as inputs for the AMF calculations. Applying the Gaussian distribution

function (GDF) described by Hong et al. [35] and Jeong et al. [36], the aerosol profile can be defined as follows:

$$\text{GDF} = \int_{z_{n1}}^{z_{n2}} W \frac{e^{-h(z-z_p)}}{[1 + e^{-h(z-z_p)}]^2} dz \quad (2)$$

$$\eta = \frac{\ln(3 + \sqrt{8})}{h} \quad (3)$$

where z_{n1} and z_{n2} are the aerosol lower and upper limits, respectively; W is a normalization constant related to total aerosol loading; h is related to the half width (HW) of aerosol vertical distribution η ; and z_p is the APH [28,35,36]. For the PBL SO_2 AMF calculation, the AOD ranges from 0.0 to 2.0 based on values obtained from the Level-3 OMI Aerosol product (L3 OMAEROe) over East Asia from 2005 to 2010. Though most aerosol particles are present near the surface, previous studies have shown that APH values of up to 2 km can exist for Asian dust cases [37–40]. APH values of 0.0, 1.0, and 2.0 km are used to investigate the effects of aerosols on the PBL SO_2 AMF according to the aerosol peak height relative to the SO_2 layer height. Values for each variable are provided in Table 1. A single scattering albedo value of 0.93 was derived from the mean value obtained from the Level-3 OMI Aerosol product (L3 OMAEROe) over East Asia for 2005–2010. The half width is fixed at 3 km, and the lower and upper limits for the aerosol layer are set to surface level and 10 km, respectively. Figure 3a shows examples of aerosol extinction profiles for various APH with AOD of 0.3. As shown in Figure 3a, for Case 1 (APH = 0 km) the aerosol layer almost overlaps with that of the SO_2 layer, but it is higher than the SO_2 layer in Cases 2, 3, and 4 (APH = 1, 2, and 3 km, respectively). For volcanic SO_2 , the AOD ranges from 0.5 to 5.0 (see Table 1) and the APH is set to 0, 4, and 8 km, which are either lower than or similar to the volcanic SO_2 layer (9–10 km; Figure 3b).

Table 1. Variables used to calculate the SO_2 AMF.

Variable		Value
O ₃ profile (TOMS climatology O ₃ profile)		High latitude, middle latitude, and low latitude
O ₃ VCD (DU)		275, 375, and 475
SZA (°)		0.1, 20, 40, 60, and 70
VZA (°)		0.1, 20, 40, 60, and 70
RAA (°)		0, 45, 90, 135, and 180
Surface Reflectance		0.0, 0.05, 0.10, 0.125, and 0.15
PBL	AOD	0, 0.3, 0.6, 0.9, 1.2, 1.5, and 2.0
	Aerosol Peak Height (km)	0, 1, 2, and 3
	SO_2 profile	From the surface to 1 km (1 km box profile)
Volcano	AOD	0.5, 1.0, 1.5, 2.0, 3.0, 4.0, and 5.0
	Aerosol Peak Height (km)	0, 4, and 8
	SO_2 profile	9–10 km (1 km box profile)

Sulfate aerosols were assumed for industrial regions with high anthropogenic SO_2 emissions, and volcanic ash was used for the volcanic SO_2 case. Model input values for each aerosol type, such as refractive index, fine-mode fraction, fine- and coarse-mode radii and variance, can be found in Torres et al. [41].

2.3. Observational Geometry and Surface Reflectance

Five SZA and VZA values (0.1°, 20°, 40°, 60°, and 70°) and five relative azimuth angle (RAA) values (0°, 45°, 90°, 135°, and 180°) were used for the SO_2 AMF calculations. According to several previous studies, SO_2 AMF is strongly dependent on the surface reflectance [10,23,26]. Thus, surface

reflectance was set to range from 0.0 to 0.15, which includes values typical of grassland, ocean, and deciduous forest in the UV–VIS range.

3. Results

3.1. Effects of Geometry and Aerosol Optical Depth on PBL SO₂

Figure 4 shows SO₂ AMF variations as a function of satellite measurement geometry (e.g., RAA, SZA, and VZA) and AOD. The SO₂ AMF is greatly affected by SZA and VZA, whereas RAA shows negligible contributions to the SO₂ AMF variation because the geometric AMF, which consists of the SZA and VZA, is enhanced as a result of the increased light path length through the Earth's atmosphere between the Sun and the space-borne sensor. The SO₂ AMF tends to decrease with increasing SZA (Figure 4a). When SZA is 20° and 60°, the SO₂ AMF at RAA of 90° is 0.23 and 0.11, respectively (Figure 4a), which suggests a shortened SO₂ absorption light path length in the PBL. This shorter length within the PBL results in fewer photons reaching the PBL for higher SZA conditions. The decrease in SO₂ AMF with increasing SZA is the inverse of the correlation between NO₂ AMF and SZA reported by Hong et al. [35]. The NO₂ AMF tends to increase with increasing SZA [35]. This difference between the trends of SO₂ and NO₂ AMFs with SZA can be attributed to the absorption wavelength of SO₂ that is generally used for spectral fitting, which is much shorter and more affected by ozone absorption and Rayleigh scattering than is that of NO₂.

Figure 4. Changes in PBL SO₂ AMF (a) as a function of the relative azimuth angle (RAA) and viewing zenith angle (VZA) at low and high solar zenith angles (SZA) under the conditions of surface reflectance = 0.05, O₃ vertical column density (VCD) = 275 DU, wavelength = 318 nm, and aerosol optical depth (AOD) = 0, and (b) as a function of RAA and AOD at low and high SZA under conditions of surface reflectance = 0.05, O₃ VCD = 275 DU, wavelength = 318 nm, and aerosol peak height (APH) = 0 km.

Figure 4b shows the effects of AOD and measurement geometry (SZA and RAA) on SO₂ AMF. The difference between SO₂ AMFs at SZA 20° and 60° is found to increase with increasing AOD, which indicates a large influence of AOD on the PBL SO₂ AMF. The SO₂ AMFs at SZA 20° and 60° are 0.33 (0.63) and 0.27 (0.53), respectively, for an AOD of 0.0 (0.6) and a RAA of 90° (Figure 4b). However, at SZA 20° and 60°, the SO₂ AMFs are 0.70 (0.72) and 0.55 (0.50), respectively, for an AOD of 0.9 (2.0) and a RAA of 90°. The SO₂ AMFs for both SZA conditions (20° and 60°) are much larger at high AOD than at low AOD. These enhanced SO₂ AMF values at high AOD may be due to the increased SO₂ absorption light path length caused by the multiple scattering effect within the PBL when SO₂

molecules and aerosols are present in the same layer (Figure 4b). However, multiple scattering caused by high aerosol loading does not necessarily lead to the increase in SO₂ AMF for certain aerosol peak heights, as discussed later in this section. In comparison with the large AOD effects under high and low SZA conditions on SO₂ AMFs, the variation of the SO₂ AMF with changes in RAA is found to be small under both high and low AOD conditions (Figure 4b). Thus, the AOD effect on the SO₂ AMF changes more with SZA than with RAA.

As shown in Figure 5a, both large SZA and large VZA lead to a significant decrease in SO₂ AMFs, and both small SZA and small VZA increase the SO₂ AMFs. This suggests that both high SZA and high VZA decrease the absorption light path length in the PBL. A small number of photons can reach the PBL at large SZA, and few of them from the PBL with the presence of SO₂ reach the satellite sensor at large VZA because of the increased photon loss at short SO₂ absorption wavelengths caused by Rayleigh scattering and stratospheric ozone absorption. Competition between the effects of ozone amount and measurement geometry on the PBL SO₂ AMF was also investigated, and is discussed later in this section. High AOD (1.2) leads to higher SO₂ AMF compared with low AOD (0.3) for most SZA and VZA conditions, except for very high VZA and SZA (>60°). The larger SO₂ AMFs for high AOD (1.2) than for small AOD (0.3) suggest that high aerosol loadings cause multiple scattering, which leads to a longer light path length in the PBL where both aerosols and SO₂ are present. However, under very large VZA and SZA (>60°), SO₂ AMFs for high AOD (1.2) are found to be slightly smaller than for low AOD (0.3). This inverse relationship of SO₂ AMF under high and low AOD conditions at high SZA and VZA is next discussed in detail using the results shown in Figure 5b.

Figure 5. Changes in PBL SO₂ AMF (a) as a function of SZA and VZA at low and high AOD, and (b) as a function of SZA and AOD under conditions of VZA = 40°, RAA = 45°, surface reflectance = 0.05, O₃ VCD = 275 DU, wavelength = 318 nm, and APH = 0 km.

The SZA and AOD effects on the SO₂ AMF were investigated in detail, as the RAA effect on the PBL SO₂ AMF is found to be small. SO₂ AMFs at large SZA tend to be smaller than those at small SZA (Figure 4a,b and Figure 5a). However, at large SZA (40°–70°), the SO₂ AMF tends to increase only to certain AOD values before it begins to decrease as AOD increases. For example, the SO₂ AMF at a SZA of 60° increases as a function of AOD over the AOD range 0.0–0.9 and tends to decrease with increasing AOD over the AOD range 0.9–2.0. Under low aerosol loading, the increased light path length that results from multiple scattering in the PBL is dominant. However, when AOD becomes high, the aerosol layer provides more of a shielding effect, which reduces the enhancement of light path length in the PBL. Even though shielding effects reduce the enhancement of the light path length under high AOD conditions, aerosols still play an enhancing role in SO₂ AMF calculations, compared with the case of no aerosols (AOD = 0). The differences between SO₂ AMFs for SZA < 40° are small compared with

those for $\text{SZA} > 40^\circ$, as there is little difference in geometric AMFs in low SZA conditions ($\text{SZA} < 40^\circ$). These small differences in geometric AMFs lead to little change in light path lengths in the PBL, which can be partly attributed to the similar geometrical AMFs between $\text{SZA} = 0.1^\circ$ and 40° . In particular, when no aerosols are present, SO_2 AMFs for low SZA ($\text{SZA} < 40^\circ$) are almost similar. However, in the presence of aerosols ($\text{AOD} > 0$), SO_2 AMFs for $\text{SZA} = 0.1^\circ$ tend to be smaller than those for $\text{SZA} = 20^\circ$ and 40° for all but the highest AOD conditions ($\text{AOD} > 1.5$). The higher SO_2 AMFs for $\text{SZA} = 0.1^\circ$ than for $\text{SZA} = 20^\circ$ or 40° in the presence of aerosols may be sensitive to aerosol composition.

3.2. Effects of Ozone Column and Aerosol Optical Depth on PBL SO_2

Figure 6 compares the ozone and SZA effects on the SO_2 AMF as a function of AOD. As discussed for the results shown in Figure 5b (Section 3.1), SO_2 AMFs for small SZA tend to be higher than those for large SZA because they are less affected by Rayleigh scattering in the troposphere and ozone absorption in the stratosphere. In terms of O_3 effects, SO_2 AMFs are found to decrease with increasing of the total O_3 amount. However, this decrease is small. For an AOD of 1.5 (0) and a SZA of 20° , the SO_2 AMFs are 0.76 (0.35), 0.75 (0.34), and 0.73 (0.33) for total ozone = 275, 375, and 475 DU, respectively. For $\text{SZA} = 60^\circ$, the SO_2 AMFs are 0.57 (0.28), 0.54 (0.27), and 0.51 (0.26) for total ozone = 275, 375, and 475 DU, respectively, for an AOD of 1.5 (0). The SO_2 AMFs are found to be more affected by SZA than by the O_3 amount, particularly over the AOD range 0.5–2.0 (Figure 6). For AOD = 0.9 (2) and total O_3 = 375 DU, the SO_2 AMFs for $\text{SZA} = 20^\circ$ and 60° are 0.72 (0.73) and 0.56 (0.50), respectively. This larger SZA effect on the SO_2 AMF compared with the O_3 effect shows greater photon loss from Rayleigh scattering than from stratospheric O_3 absorption. The short SO_2 absorption wavelength coupled with high SZA leads to enhanced Rayleigh scattering and stratospheric O_3 absorption before photons reach the PBL where SO_2 molecules are present. More photons reach the PBL under low SZA conditions than under high SZA conditions, where they undergo multiple scattering within the PBL, causing the SO_2 absorption light path length to increase significantly compared with high SZA conditions. Although ozone effects on the SO_2 AMF are smaller than those of SZA and AOD, the effect of the aerosol vertical profile is examined next.

Figure 6. (a) Changes in PBL SO_2 AMF as a function of AOD under various total O_3 column densities, and (b) O_3 profiles at low and high SZA ($\text{VZA} = 40^\circ$; $\text{RAA} = 45^\circ$; surface reflectance = 0.05; wavelength = 318 nm; $\text{APH} = 0$ km).

3.3. Effects of Aerosol Peak Height and Aerosol Optical Depth on PBL SO_2

As discussed for the results shown in Figures 4 and 5, aerosols increase the light path length and the PBL SO_2 AMF by multiple scattering effects when aerosols and SO_2 are present in the same layer. We investigated changes in the aerosol effects on the SO_2 AMF with changes in the aerosol peak height relative to the SO_2 layer height. The vertical profiles of SO_2 and aerosols, with indicators of their corresponding layers, are shown in Figure 3. Figure 7a shows the APH effect on the SO_2 AMF as a function of AOD. The PBL SO_2 AMF is found to increase with decreasing APH, which indicates an increase in light path length in the PBL caused by a large multiple scattering effect when both the

aerosol and SO₂ layers are at the same height. For AOD values of 0.3 (2.0), the SO₂ AMF is 0.55 (0.75), 0.36 (0.21), and 0.28 (0.10) for an APH of 0, 1, and 2 km, respectively. For APH = 0 km (Case 1 in Figure 3a) where the aerosol layer almost overlaps the SO₂ layer, the AOD increases the SO₂ AMF. The SO₂ AMF increases from 0.55 to 0.75 with an increase in AOD from 0.3 to 2.0. However, aerosols are found to decrease the SO₂ AMF for APH = 1, 2, and 3 km (Cases 2, 3, and 4 in Figure 3a). For APH = 2 and 3 km where the aerosol layer is located above that of SO₂, the SO₂ AMF tends to decrease with increasing AOD, which suggests an aerosol shielding effect. The shielding effect is found to be more important than the multiple scattering effect for APH = 1 km where the aerosol layer is slightly higher than but partially overlaps that of SO₂. For APH = 1 km, the SO₂ AMF is 0.36 and 0.21 for an AOD of 0.3 and 2.0, respectively, which shows the decreasing trend of SO₂ AMF as a function of AOD. The APH effect on SO₂ AMF is also investigated with respect to both SZA and AOD. The trend of SO₂ AMF as a function of APH and AOD (Figure 7b) is similar to those shown in Figure 7a. However, the SO₂ AMF for high SZA (SZA = 60°) is found to be smaller than for low SZA (SZA = 20°) for all APH and AOD conditions because of greater Rayleigh scattering under high SZA conditions, as discussed for the results shown in Figures 4–6.

Figure 7. (a) Changes in PBL SO₂ AMF as a function of AOD for various APH, and (b) simultaneous effects of AOD and APH at low and high SZA (VZA = 40°; RAA = 45°; surface reflectance = 0.05; O₃ VCD = 275 DU; wavelength = 318 nm; APH = 0 km).

3.4. Effects of Aerosol Optical Depth and Surface Reflectance on PBL SO₂

Surface reflectance effects on SO₂ AMF are also investigated. As SZA, AOD, and APH are found to have important effects on SO₂ AMF (Figures 4–7), the surface reflectance effect is quantified as a function of AOD and SZA for APH = 0 and 2 km. Surface reflectance is found to increase the SO₂ AMF. When SO₂ and aerosols are present in the same layer (APH = 0 km), SO₂ AMF tends to increase significantly with increasing surface reflectance under low aerosol loading conditions (Figure 8). For SZA = 60° (20°), the SO₂ AMF increases from 0.41 (0.43) to 0.68 (0.81) as surface reflectance increases from 0.0 to 0.15 for AOD = 0.3, but increases only slightly from 0.52 (0.72) to 0.56 (0.81) for AOD = 2.0, indicating a large effect of surface reflectance on the PBL SO₂ AMF under low aerosol conditions.

When the aerosol layer is higher than the SO₂ layer (APH = 2 km), it has a shielding effect and the SO₂ AMF tends to increase significantly with increasing surface reflectance, particularly under low AOD conditions. For SZA = 60° (20°) and APH = 2 km, the SO₂ AMF increases from 0.11 (0.15) to 0.37 (0.55) for AOD = 0.3 as surface reflectance increases from 0.0 to 0.15, increasing by a factor of greater than three. The SO₂ AMF increases from 0.02 (0.04) to 0.07 (0.13) for AOD = 1.8 and SZA = 60° (20°). Under high AOD conditions for both APH = 0 and 2 km, surface reflectance affects the PBL SO₂ AMF less than it does under low AOD conditions. Thus, the PBL SO₂ AMF is found to be affected more by SZA, VZA, AOD, APH, and surface reflectance than by RAA, total column O₃, and the O₃ vertical profile.

Figure 8. Changes in PBL SO_2 AMF as a function of AOD and surface reflectance at APH = 0 and 2 km under the conditions of (a) $\text{SZA} = 20^\circ$ and (b) $\text{SZA} = 60^\circ$ ($\text{VZA} = 40^\circ$; $\text{RAA} = 45^\circ$; surface reflectance = 0.05; O_3 VCD = 275 DU; wavelength = 318 nm).

3.5. Effects of Aerosol Peak Height and Aerosol Optical Depth on Volcanic SO_2

We also investigated the AMF for volcanic SO_2 , which is located at a much higher altitude than is the case for PBL SO_2 . Figure 9 shows the effects of SZA, AOD, and APH on volcanic SO_2 AMF. Here, we use a 10 km SO_2 layer height [34] and volcanic aerosols [41] to calculate the volcanic SO_2 AMF. The APH is set to 0, 4, and 8 km, which are either lower than or close to the volcanic SO_2 layer between 9 and 10 km (Figure 3b). The volcanic SO_2 AMF is found to be significantly affected by SZA. However, SZA affects the volcanic SO_2 AMF differently than the PBL SO_2 AMF. High SZA is found to increase the volcanic SO_2 AMF (Figure 9), whereas the PBL SO_2 AMF decreases with increasing SZA (Figure 8). This increasing trend of volcanic SO_2 AMF with increasing SZA suggests a weaker Rayleigh scattering effect at high SZA at altitudes above the PBL. Higher SZA increases the light path length within the volcanic SO_2 layer because of decreased photon loss. These photons penetrate into the SO_2 layer (above the PBL), which leads to higher volcanic SO_2 AMFs.

Figure 9. Changes in volcanic SO_2 AMF as a function of AOD and APH at low and high SZA ($\text{VZA} = 40^\circ$; $\text{RAA} = 45^\circ$; surface reflectance = 0.05; O_3 VCD = 275 DU; wavelength = 375 nm; aerosol type = volcanic ash).

For $\text{SZA} = 60^\circ$, the volcanic SO_2 AMFs tend to decrease with increasing APH under high AOD conditions. For $\text{AOD} = 5.0$, the volcanic SO_2 AMFs are 3.73 and 3.40 for $\text{APH} = 0$ and 8 km, respectively. Under very high AOD conditions, the aerosol layer under high aerosol loading conditions at an APH of 8 km completely overlaps the volcanic SO_2 layer, which leads to a large shielding effect. The SO_2 AMF is much lower at $\text{SZA} = 20^\circ$ than that at $\text{SZA} = 60^\circ$. Lower SZA reduces the light path length within the volcanic SO_2 layer. For low AOD (0–2) and $\text{SZA} = 60^\circ$, the volcanic SO_2 AMF is not significantly affected by APH.

3.6. Estimation of SO_2 AMF Errors

3.6.1. PBL SO_2 AMF Error

SO_2 AMF errors can be caused by inaccuracy of the input data used for the RTM calculations. The AMF errors are quantified as a function of uncertainty in the input data for AOD, APH, and surface reflectance, as these affect the SO_2 AMF the most among parameters. An APH of 1 km, an AOD of 0.27, and a surface reflectance of 0.05 were used as ‘true’ values for the PBL SO_2 AMF calculation. In Figures 10 and 11, true AOD and surface reflectance values were obtained from mean values obtained from the Level-3 OMI Aerosol product (OMAEROe) and Level-3 OMI LER climatology (OMLER), respectively, over East Asia from 2005–2010. The uncertainties in AOD and surface reflectance input data were based on the MODIS Near Real Time (NRT) product (MYD04 and MYD09, respectively). Detailed uncertainty information is provided in Table 2. The expected error of APH is assumed as 1 km, which is suggested in Fishman et al. [42].

Figure 10. Percent difference in PBL SO_2 AMF calculated with various AOD, APH, and SZA values and compared with a ‘true’ AMF that is calculated for $\text{AOD} = 0.27$, $\text{APH} = 1$ km, and $\text{SZA} = 20^\circ$ and 60° ($\text{VZA} = 40^\circ$; $\text{RAA} = 0^\circ$; surface reflectance = 0.05; O_3 VCD = 275 DU; wavelength = 318 nm; aerosol type = sulfate).

Figure 11. Percent difference in PBL SO₂ AMF calculated with various AOD, surface reflectance, and APH values compared with a ‘true’ AMF that is calculated for AOD = 0.27, surface reflectance = 0.05, and APH = 0 and 1 km for (a) SZA = 20° and (b) SZA = 60° (VZA = 40°; RAA = 0°; O₃ VCD = 275 DU; wavelength = 318 nm; aerosol type = sulfate).

Table 2. AOD and surface reflectance uncertainty.

Input Data	Uncertainty	Reference
MODIS AOD (MYD04)	$\pm (0.05 + 0.15 \times \text{AOD})$	Chu et al. [43]
MODIS Surface reflectance (MYD09)	$\pm (0.005 + 0.05 \times \text{surface reflectance})$	EOS Land Validation: https://landval.gsfc.nasa.gov
Aerosol height	± 1 km (expected error)	Fishman et al. [42]

Figure 10 shows the percent difference in PBL SO₂ AMF as a function of APH and AOD against the true AMF. The VZA and RAA were set to 40° and 0°, respectively, and two SZA conditions (20° and 60°) were used. The true AMF values, which are calculated for APH = 1 km and AOD = 0.27, are 0.36 and 0.28 for SZA = 20° and 60°, respectively (Figure 10). Under high SZA conditions, the PBL SO₂ AMFs are more strongly affected by AOD and APH than under low SZA conditions. In particular, when the APH used in the AMF calculation is smaller than the ‘true’ APH, the SO₂ AMF error is significantly larger under high SZA conditions. When APH = 0 km (1 km smaller than the ‘true’ value) and AOD = 0.18 (0.33% smaller than the ‘true’ AOD of 0.27), the SO₂ AMF errors are +60% and +37% for SZA = 60° and 20°, respectively (Figure 10). When APH = 0 km and AOD = 0.36 (0.33% larger than the ‘true’ AOD), the SO₂ AMF error is overestimated by 88% (63%) for SZA 60° (20°).

When the APH used in the AMF calculation is larger than the ‘true’ APH, the magnitude of the SO₂ AMF error is found to be smaller than when the APH that is used is smaller than the ‘true’ APH. When an artificially large APH value (2.0 km) is used, the SO₂ AMF error is underestimated by 21% (15%) for SZA = 60° (20°) with AOD = 0.18 (33% smaller than the ‘true’ AOD). When APH = 2 km and AOD = 0.36 (33% larger than the ‘true’ AOD), the SO₂ AMF errors are −37% and −26% for SZA = 60° and 20°, respectively.

We also quantified the contributions of surface reflectance uncertainties to the PBL SO₂ AMF error. Figure 11 shows the percent difference in PBL SO₂ AMF as a function of surface reflectance and AOD against the ‘true’ AMF. The ‘true’ PBL SO₂ AMF (AMF error = 0 %) is calculated for AOD = 1.0 and APH = 4 km. The PBL SO₂ AMFs under high SZA conditions are found to be more affected by AOD and surface reflectance than those under low SZA conditions. In addition, the trend of SO₂ AMF as a function of AOD and surface reflectance for SZA = 20° is opposite that when SZA = 60°. When surface reflectance = 0.042 (15% smaller than the ‘true’ value) and AOD = 0.18 (33% smaller than the ‘true’ AOD of 0.27), the SO₂ AMF error is underestimated by 15% and 3% for APH = 0 and 2 km, respectively, and SZA = 20° (Figure 11a). However, under the same surface reflectance (0.042) and AOD (0.18)

conditions but for SZA = 60°, the SO₂ AMF error is 3% and −13% for APH = 0 and 2 km, respectively (Figure 11b). For a surface reflectance of 0.057 (15% larger than the ‘true’ value) and AOD = 0.18, the SO₂ AMF error is −4% and 18% for APH = 0 and 2 km, respectively, and SZA = 20° (Figure 11a). For the same surface reflectance (0.0575) and AOD (0.18) conditions but with SZA = 60°, the SO₂ AMF error is 23% and −5% for APH = 0 and 2 km, respectively (Figure 11b). The PBL SO₂ AMF error is also found to be both negatively and positively biased, depending on the AOD, APH, and surface reflectance error. For example, for high APH (APH = 2 km), overestimated AOD (AOD = 0.36) leading to aerosol shielding effects, and a surface reflectance of 0.042, the SO₂ AMF error is calculated to be −16%, which is negatively biased compared with the ‘true’ AMF. This indicates that a positively biased AOD value under high APH conditions (APH = 2 km) artificially increases aerosol shielding effects and leads to a SO₂ AMF value that is smaller than the ‘true’ value. This smaller SO₂ AMF is, thus, negatively biased.

The error budget of the PBL SO₂ AMF is additionally calculated based on the TROPOMI measurement condition, since the TROPOMI is the most recent sensor that monitors SO₂ column densities. The PBL SO₂ AMF error budget was calculated on the basis of the uncertainties associated with the APH, AOD, and surface reflectance because we found that the SO₂ AMF errors were caused mainly by the APH, AOD, and surface reflectance rather than the measurement geometries and ozone profile. We used Gaussian error propagation (GEP) to estimate the PBL SO₂ AMF error as follows [44,45].

$$\sqrt{\left(\frac{\partial \text{AMF}}{\partial \chi_i}\right)^2 \sigma_{\chi_i}^2} = \varepsilon_{\text{AMF}, \chi_i} \quad (4)$$

$$\varepsilon_{\text{AMF}} = \sqrt{\varepsilon_{\text{AMF}, \text{AOD}}^2 + \varepsilon_{\text{AMF}, \text{surface reflectance}}^2 + \varepsilon_{\text{AMF}, \text{APH}}^2} = \sqrt{\sum_{i=1}^3 \left(\frac{\partial \text{AMF}}{\partial \chi_i}\right)^2 \sigma_{\chi_i}^2} \quad (5)$$

where $\varepsilon_{\text{AMF}, \chi_i}$ is the error of the PBL SO₂ AMF caused by the input parameters χ_i , ε_{AMF} is the total error of the PBL SO₂ AMF, $\left(\frac{\partial \text{AMF}}{\partial \chi_i}\right)$ is the partial derivative of the PBL SO₂ AMF by the input parameters χ_i , σ_{χ_i} represents the uncertainty of the i^{th} input parameter χ_i , and $\sigma_{\chi_i}^2$ is the variance. Equation (5) assumes that errors from the various input parameters are independent of one another. We calculated the error budget of the PBL SO₂ AMF error as the percentage error to effectively present the SO₂ AMF error caused by uncertainties in APH, AOD, and surface reflectance (Equations (6) and (7)). To calculate the percentage error for each parameter, $\varepsilon_{\text{AMF}, \chi_i}$ was divided by the true PBL SO₂ AMF and then averaged using the number of values in dataset z (Equation (6)). To calculate the total percentage error, ε_{AMF} was divided by the true PBL SO₂ AMF and then averaged using the number of values in dataset z (Equation (7)).

$$\text{percentage error for } \chi_i (\%) = \frac{\sum_0^z \frac{\varepsilon_{\text{AMF}, \chi_i}}{\text{True SO}_2 \text{ AMF}}}{z} \times 100 \quad (6)$$

$$\text{total percentage error } (\%) = \frac{\sum_0^z \frac{\varepsilon_{\text{AMF}}}{\text{True SO}_2 \text{ AMF}}}{z} \times 100 \quad (7)$$

According to the TROPOMI SO₂ Algorithm Theoretical Basis Documents (ATBD), the climatological monthly minimum Lambertian equivalent reflector (minLER) data from Kleipool et al. [46] are used as the input data for surface reflectance during operational SO₂ retrieval [47]. We used an uncertainty of 0.02 for the minLER data in our error budget calculations [47]. Aerosol parameters, such as the aerosol extinction profile, which accounts for the AOD and APH, are not considered in the operational retrieval of TROPOMI SO₂ [47]. Therefore, in our error budget calculations, we assumed that the uncertainty associated with the MODIS AOD was the same as the AOD for TROPOMI SO₂ retrieval [43]. The uncertainty associated with the APH was assumed to be 1 km according to the previous study [42]. To calculate $\left(\frac{\partial \text{AMF}}{\partial \chi_i}\right)^2$, we simulated the PBL SO₂ AMF using an RTM with input parameters to assume the measurement condition of the TROPOMI as shown in

Table 3. To calculate the error budget in thin and thick aerosol cases, two AOD intervals (interval A: $0.2 \leq \text{AOD} \leq 1.0$; interval B: $1.0 < \text{AOD} \leq 2.0$) were considered. In addition, the SO_2 AMF was found to be affected by the APH relative to the SO_2 layer height, and three APH conditions (0, 1, and 2 km) were considered in our error analysis. For the APH, AOD, and surface reflectance, the new SO_2 AMF was simulated using $X_i + \sigma_{X_i}$. ∂X_i denotes the difference between the true X_i and $X_i + \sigma_{X_i}$, and ∂AMF is the difference between the ‘true’ AMF simulated with ‘true’ input values and the new AMF simulated using input parameters, with the uncertainty of each parameter being $X_i + \sigma_{X_i}$.

Table 3. Variables and values used in calculating the PBL SO_2 AMF error budget related to uncertainties in the APH, AOD, and surface reflectance based on the TROPOMI measurements.

Variable	Value
SZA (°)	10, 30, 50, and 70
VZA (°)	10, 30, 50, and 70
RAA (°)	0.0, 45.0, 90.0, 135.0, and 180.0
surface reflectance	0.05, 0.10, 0.15, and 0.20
aerosol optical depth	Interval A: 0.2, 0.4, 0.6, 0.8, and 1.0 Interval B: 1.2, 1.4, 1.6, 1.8, and 2.0
aerosol peak height [km]	0, 1, and 2
O_3 profile (TOMS climatology O_3 profile)	Middle latitude
O_3 VCD (DU)	275 DU
PBL SO_2 profile	From the surface to 1 km (1 km box profile)

Table 4 summarizes the total error budget of the PBL SO_2 AMF caused by the uncertainties associated with the individual input parameters (APH, AOD, and surface reflectance) and the TROPOMI measurement conditions using Equations (6) and (7). In general, the APH uncertainty makes the greatest contribution to the total SO_2 AMF error among the individual input parameters based on the TROPOMI measurement conditions (Table 3). In particular, when the APH is 1 km, inaccurate APH values can lead to PBL SO_2 AMF percentage errors of 72.1% under lower AOD conditions (interval A), whereas the PBL SO_2 AMF error related to APH uncertainty increases to 173.4% under higher AOD conditions (interval B). It is also evident that uncertainties in APH and AOD tend to cause higher PBL SO_2 AMF percentage errors for high AODs (interval B) compared with low AODs (interval A), which suggests that the uncertainties in APH and AOD have a greater effect on the PBL SO_2 AMF percentage error under high AOD conditions. However, for high SO_2 conditions, the effect of the surface reflectance uncertainty on the PBL SO_2 AMF is smaller than that of the AOD and APH in the error budget, based on the TROPOMI measurement conditions.

Table 4. Summary of PBL SO_2 AMF errors caused by uncertainties in APH, AOD, and surface reflectance based on the TROPOMI measurements.

	$0.2 \leq \text{AOD} < 1.0$	$1.0 \leq \text{AOD} < 2.0$
APH = 0 km	SO_2 AMF percentage error (%)	SO_2 AMF percentage error (%)
AOD	3.8%	4.2%
Surface reflectance	6.4%	2.8%
APH	49.3%	77.3%
Total error	49.9%	77.5%
APH = 1.0 km	SO_2 AMF percentage error (%)	SO_2 AMF percentage error (%)
AOD	9.1%	21.0%
Surface reflectance	12.5%	9.1%
APH	70.5%	171.9%
Total error	72.1%	173.4%
APH = 2.0 km	SO_2 AMF percentage error (%)	SO_2 AMF percentage error (%)
AOD	19.1%	42.2%
Surface reflectance	19.4%	20.2%
APH	37.1%	110.8%
Total error	46.0%	120.2%

3.6.2. Volcanic SO₂ AMF Error

Volcanic SO₂ AMF errors are also quantified as a function of the uncertainties of APH and AOD, which are used as inputs for the RTM calculations. Figure 12 shows the percent difference in volcanic SO₂ AMF as a function of APH and AOD against the ‘true’ AMF. To calculate the ‘true’ volcanic SO₂ AMF, AOD = 1.0 and APH = 4 km were used following previous studies [48–51] that reported volcanic plume heights during the Eyjafjallajökull eruptions. Volcanic SO₂ AMF errors for a SZA of 20° are similar to those for SZA = 60° when APH = 0 km, instead of the ‘true’ APH of 4 km. The volcanic SO₂ AMF errors are −3% (−4%) and 12% (10%) for AOD values of 0 and 5, respectively, when the ‘true’ AOD is 1. However, volcanic SO₂ AMF errors for SZA = 20° increase significantly when APH = 8 km and AOD = 0 (for a ‘true’ APH of 4 km and AOD of 1). The volcanic SO₂ AMF error for SZA = 20° is 13% for APH = 8 km and AOD = 5. This positively biased volcanic SO₂ AMF may be associated with enhanced multiple scattering effects for the artificially high APH value of 8 km for SZA = 20°.

Figure 12. Percent difference in volcanic SO₂ AMF calculated with various AOD, APH, and SZA values compared with a ‘true’ AMF that is calculated for AOD = 1.0, APH = 4 km, and SZA = 20° and 60° (VZA = 40°; RAA = 0°; surface reflectance = 0.05; O₃ VCD = 275 DU; wavelength = 375 nm; aerosol type = volcanic aerosol).

4. Discussion

The uncertainties associated with the SO₂ AMF are an important aspect of our understanding of SO₂ VCD retrieval errors generated by hyperspectral UV sensors. Previous studies [6,10,23,25,26] have reported that several factors can cause these SO₂ AMF sensitivities and uncertainties. Khokhar et al. [6] and Richter et al. [10] investigated the effect of various parameters, such as SZA, SO₂ profile, and surface reflectance, on the volcanic SO₂ AMF. Theys et al. [25] studied the SO₂ vertical distribution effect on the volcanic and PBL SO₂ AMF. Lee et al. [26] carried out error analysis of the PBL SO₂ AMF for surface reflectance, aerosol properties, cloud fraction, and the SO₂ vertical distribution. However, none of these studies investigated the critical role of APH on the SO₂ AMF under various measurement conditions. Hong et al. [35] reported the simultaneous effects of APH and other aerosol properties on the NO₂ AMF. In this section, we discuss several important findings with respect to the APH effects on the SO₂ AMF and compare our findings with those for NO₂ AMF reported by Hong et al. [35].

The AOD effect on the PBL SO₂ AMF changes with the APH relative to the SO₂ layer height. With the existence of both SO₂ and aerosols within the PBL, the PBL SO₂ AMFs are largely increased under high-AOD conditions due to the increased SO₂ absorption caused by multiple scattering. When the aerosol layer is located above the SO₂ layer, the aerosol shielding effect leads to a reduction in the SO₂ AMF with increasing AOD. A previous study by Hong et al. [35] reported similar patterns between the NO₂ AMF and APH. The increase in AOD leads to increases in NO₂ AMF when the APH and NO₂ layer are at the same height, and to decreases in NO₂ AMF with the shielding effect due to high-APH conditions [35].

In terms of the APH effect on the volcanic SO₂ AMF, under very high AOD and SZA, APH has a significant influence on the volcanic SO₂ AMF, which is reduced by the shielding effect. The APH also influences the PBL SO₂ AMF, which can either be reduced or increased depending on the AOD condition and APH relative to the SO₂ layer height.

Increases in the SZA and VZA cause the PBL SO₂ AMF to decrease. The Rayleigh scattering effect generated at high SZA or VZA values is found to largely decrease the light path length at the SO₂ absorption wavelength. The decrease in SO₂ AMF with increasing SZA is the inverse of the correlation between NO₂ AMF and SZA reported by Hong et al. [35]. This difference between the trends of SO₂ and NO₂ AMFs with SZA can be attributed to the absorption wavelength of SO₂ that is generally used for spectral fitting, which is much shorter and more affected by ozone absorption and Rayleigh scattering than is that of NO₂.

Hence, in this present study, we additionally calculated the error budget of the PBL SO₂ AMF caused by uncertainties associated with the APH, AOD, and surface reflectance based on the TROPOMI measurement conditions. The APH uncertainty was the most important factor with respect to the PBL SO₂ AMF error (up to 171.9%). Uncertainties in both the APH and AOD tend to cause an increase in the PBL SO₂ AMF percentage error under high AOD conditions and when using the TROPOMI measurement conditions. However, the effect of the uncertainty related to the surface reflectance on the SO₂ AMF error is not sensitive to the AOD level. In this present study, the high SO₂ AMF error caused by APH uncertainty in the error budget shows that aerosol height needs to be considered in the error budget of operational SO₂ retrieval algorithms.

In the future, the SO₂ AMF sensitivities and error analysis provided here could be used to inform error reduction strategies for an SO₂ retrieval algorithm developed for use with new space-borne satellite sensors such as the Geostationary Environmental Monitoring Spectrometer (GEMS). Here, we have investigated the effect of APH under various measurement conditions on SO₂ AMF for clear sky conditions. In future studies, the sensitivity of SO₂ AMF to APH under cloudy conditions needs to be investigated because space-borne measurements are often performed in cloudy skies.

5. Summary and Conclusions

SO₂ retrieval error includes errors in spectral fitting and errors in the calculation of AMFs, which are used in DOAS SO₂ retrieval algorithms. Here, we examined the simultaneous effects of AOD, APH, ozone column density, ozone vertical profile, and surface reflectance under various measurement geometries (SZA, VZA, and RAA). These effects are quantified for PBL and volcanic SO₂ AMFs. The important findings of this work are summarized as follows.

The PBL SO₂ AMF tends to decrease with increasing SZA and VZA. A large Rayleigh scattering effect at high SZA or VZA is found to reduce the light path length at the SO₂ absorption wavelength used for spectral fitting. However, under high AOD conditions, the aerosol layer provides more of a shielding effect, which reduces the light path length in the PBL and, thus, the SO₂ AMF.

The influence of AOD on the PBL SO₂ AMF varies with the APH relative to the SO₂ layer height. When both SO₂ and most aerosols are present within the PBL, the PBL SO₂ AMFs are greatly enhanced under high AOD conditions because of the increased SO₂ absorption caused by the longer light path length induced by multiple scattering. When the aerosol layer is above the SO₂ layer, the SO₂ AMF tends to decrease with increasing AOD because of the aerosol shielding effect.

We found a large contribution of surface reflectance to the PBL SO₂ AMF under low aerosol and SZA conditions. Enhanced surface reflectance significantly increases the PBL SO₂ AMF.

SZA affects the volcanic SO₂ AMF differently to the PBL SO₂ AMF. Higher SZA is found to increase the volcanic SO₂ AMF because of negligible Rayleigh scattering at high altitudes. When the aerosol layer overlaps that of SO₂ under high SZA and AOD conditions, aerosol shielding effects slightly decrease the volcanic SO₂ AMF.

Author Contributions: H.L. designed and interpreted the entire experiments. W.C. simulated synthetic radiance using radiative transfer model and estimated SO₂ AMF errors. J.Y. produced input data for radiative transfer model and calculated aerosol effect on SO₂ AMF. M.V.R. interpreted results of radiative transfer calculation and summarized previous studies. J.-H.K. displayed the experimental results and compared them with results from previous studies. J.P. and D.K. calculated the effect of ozone column and surface reflectance on SO₂ AMF. All authors have read and agreed to the published version of the manuscript.

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