1	Profiling transboundary aerosols over Taiwan and assessing their radiative				
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23 Abstract

24 A synergistic process was developed to study the vertical distributions of aerosol optical 25 properties and their effects on solar heating using data retrieved from ground-based radiation 26 measurements and radiative transfer simulations. Continuous MPLNET and AERONET 27 observations were made at a rural site in northern Taiwan from 2005 to 2007. The aerosol 28 vertical extinction profiles retrieved from ground-based lidar measurements were categorized 29 into near-surface, mixed, and two-layer transport types, representing 76% of all cases. 30 Fine-mode (Ångström exponent, α , ~1.4) and moderate-absorbing aerosols (columnar 31 single-scattering albedo ~0.93, asymmetry factor ~0.73 at 440 nm wavelength) dominated in 32 this region. The column-integrated aerosol optical thickness at 500 nm (τ_{500nm}) ranges from 33 0.1 to 0.6 for the near-surface transport type, but can be doubled in the presence of 34 upper-layer aerosol transport. We utilize aerosol radiative efficiency (ARE; the impact on 35 solar radiation per unit change of τ_{500nm}) to quantify the radiative effects due to different 36 vertical distributions of aerosols. Our results show that the ARE at the top-of-atmosphere (-23 W m⁻²) is weakly sensitive to aerosol vertical distributions confined in the lower troposphere. 37 On the other hand, values of the ARE at the surface are -44.3, -40.6 and -39.7 W m⁻² for 38 39 near-surface, mixed, and two-layer transport types, respectively. Further analyses show that 40 the impact of aerosols on the vertical profile of solar heating is larger for the near-surface 41 transport type than that of two-layer transport type. The impacts of aerosol on the surface 42 radiation and the solar heating profiles have implications for the stability and convection in 43 the lower troposphere.

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INDEX TERMS: 0305 Atmospheric Composition and Structure: Aerosols and particles; 0360
 Atmospheric Composition and Structure: Transmission and scattering of radiation; 1637

Global Change: Regional climate change; 3359 Atmospheric Processes: Radiative processes;
0345 Atmospheric Composition and Structure: Pollution: urban and regional (0305, 0478, 4251) *KEYWORDS*: aerosol vertical distribution, aerosol optical properties, direct aerosol radiative effect

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55 1. Introduction

Aerosols affect the Earth's energy budget by scattering and absorbing radiation (the "direct effect") and by modifying the life cycle and properties of clouds (the "indirect effect"). The complex spatial, temporal, chemical composition, physical size and shape, as well as the optical characteristics of the atmospheric aerosols cause large uncertainties in the estimation of the effects of aerosols on climate [*IPCC*, 2007; *CCSP*, 2009].

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62 Information on the vertical distribution and the optical properties of tropospheric 63 aerosols is required for radiative transfer calculations and is of paramount importance in 64 understanding the effects of aerosols on climate [e.g., Kaufman et al., 1997a; Haywood and 65 *Ramaswamy*, 1998 and references therein]. To date, only a few recent studies have taken into 66 consideration the detailed aerosol vertical distribution that was retrieved using 67 remote-sensing techniques in estimating the radiative effects of aerosols [e.g., Johnson et al., 68 2008; McFarlane et al., 2009]. However, the often observed multi-layer aerosol transport 69 over East Asia [e.g., Muravama et al., 2004; Chiang et al., 2007] can cause uncertainties in 70 the estimation of aerosol effects on the regional climate.

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Several studies have demonstrated the ability to retrieve the vertical profiles of aerosol
 extinction using a combination of Micro Pulse Lidar (MPL) and Sun/sky radiometer

74 measurements of aerosol optical thickness (τ) [e.g., Welton et al., 2000; Welton et al., 2002; 75 Campbell et al., 2003; Schmid et al., 2006; Hayasaka et al., 2007; He et al., 2008]. The 76 NASA Micro Pulse Lidar Network (MPLNET) [Welton et al., 2001] provides coordinated 77 and standardized observations of aerosol vertical distribution from a federated network of 78 MPL systems collocated with the NASA Aerosol Robotic Network (AERONET) [Holben et 79 al., 1998] Sun/sky radiometers. However, at the present time only a subset of all MPLNET 80 sites have collected more than a few continuous years of data. Thus, long-term studies of 81 MPLNET derived aerosol vertical distribution are only just now becoming possible. In this 82 paper, we first demonstrate a long-term database obtained from an AERONET and MPLNET 83 collocated site to study aerosol vertical distributions of optical properties. The data 84 acquisition rate is used to evaluate the state-of-the-art measurement strategy for this site.

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86 In this study, we present a synergistic process to characterize the vertical distributions of 87 aerosols, their optical properties, and the direct aerosol radiative effect (DARE) using 88 ground-based remote-sensing and a radiative transfer model. Three years (from 2005 to 2007) of vertical aerosol extinction (σ , km⁻¹) profiles and column-integrated aerosol optical 89 90 properties were derived from lidar and Sun/sky radiometers measurements at a rural location 91 in Taiwan. We categorized the aerosol extinction profiles into three types, representing the 92 major characteristics of the vertical distribution of aerosol over Taiwan. The integration with 93 trajectories analysis, space-based remote-sensing and radiative transfer calculations provided 94 insights into understanding the relationships between the vertical distribution of aerosol, the 95 transport mechanism, its optical properties and its radiation effects.

97 2. Measurements and Data Usage

98 2.1. Site Description

99 The aerosol data used in this study were taken from observations at National Central 100 University (NCU) in Chungli City (24.97°N, 121.18°E; 133 m above sea level), 50 km south 101 of Taipei, Taiwan. Chungli is a medium-sized city with a population of ~360,000. NCU 102 serves as a rural site because it is located at the western edge of Chungli with no significant 103 emission sources nearby. The meteorological conditions are characterized by the 104 southwesterly Asian monsoon in the summer and the northeasterly monsoon in the winter. 105 The weather is humid and cloudy during the summer, but dry and relatively clear during the 106 winter. Studies have shown that northern Taiwan is located on the pathway of the pollution 107 transport from Asia to the Pacific Ocean during pollution outbreaks [Liu et al., 2006; Chiang 108 *et al.*, 2007].

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110 **2.2.** Sun/sky radiometer

111 The CIMEL Electronique CE-318 Sun/sky radiometer measurements reported in this 112 paper were made by instruments that participate in AERONET [Holben et al., 1998]. A 113 brief description of the instrumentation and its calibration can be found on the AERONET 114 website (http://aeronet.gsfc.nasa.gov/). The Sun/sky radiometer makes direct solar irradiance 115 measurements with a 1.2° full field of view every 15 min at 340, 380, 440, 500, 675, 870, 940, 116 and 1020 nm (nominal wavelength, λ). The uncertainty of the measured τ , due primarily to 117 the calibration uncertainty, is ± 0.01 for $\lambda > 440$ nm and ± 0.02 for $\lambda \le 440$ nm. In addition to the 118 direct solar irradiance measurements, the Sun\sky radiometer measures the sky radiance at 119 four wavelengths (440, 675, 870, and 1020 nm) along the solar almucantar (i.e., at a constant

120 elevation angle, with varied azimuth angles) up to eight times a day. The sky radiance 121 measurements are used to retrieve additional columnar aerosol properties including volume 122 size distribution, phase function, real and imaginary components of the refractive index, 123 effective particle radius, single-scattering albedo (ω), and the asymmetry factor (g), which are 124 routinely computed with the AERONET inversion algorithms [Dubovik and King, 2000; 125 Dubovik et al., 2006; Holben et al., 2006]. Specifically, the retrievals of ω and the imaginary 126 part of the refractive index are further limited to $\tau_{440nm} \ge 0.4$ due to an increased uncertainty 127 in the absorption properties of the inversion retrieval during a lower aerosol loading in the 128 atmosphere [Dubovik et al. 2002a and 2002b]. The uncertainty in the retrieved ω is estimated 129 to be ± 0.03 .

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131 Two Sun/sky radiometers are located at the NCU campus. One of them is jointly 132 operated by the Taiwan Environmental Protection Administration (EPA) and the Department 133 of Atmospheric Sciences, NCU (hereafter referred to as the EPA-NCU site). The other 134 radiometer, located within ~200 m of the EPA-NCU site, is operated by the Center for Space 135 and Remote Sensing Research, NCU (hereafter referred to as the NCU_Taiwan site).

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137 2.3. Micro Pulse Lidar (MPL)

The EPA-NCU MPL system is a member of the NASA MPLNET project (http://mplnet.gsfc.nasa.gov/). The MPL [*Spinhirne et al.*, 1995] is a compact and eye-safe single wavelength (527 nm) elastic backscatter lidar system capable of determining the range of both aerosols and clouds by firing a short pulse of laser light and measuring the time-of-flight from the pulse transmission to the reception of the returned signal. Our system 143 supports long-term measurements with 1-minute time resolution and 0.075 km vertical 144 resolution. The standard instrument design and level 1 signal processing are described in 145 detail by Campbell et al. [2002] and Welton et al. [2002]. Real time data products (level 1.5, 146 no quality assurance) are provided on a next day basis, and include identification of multiple 147 cloud layer heights (base and top), the planetary boundary layer (PBL) height, and the height 148 of the highest aerosol layer. Time resolutions for each product are: clouds (1 minute), aerosols 149 (20 minutes), and PBL (5 minutes). The derivation of level 1.5 aerosol properties is based on 150 the algorithm by Welton et al. [2000] where the AERONET level 1.5 aerosol optical thickness 151 is used as the constraint to solve for the lidar ratio and the extinction and optical thickness 152 profiles from the cloud screened 20-minute signal averages. The mean uncertainty in MPLNET retrieved extinction is ± 0.015 km⁻¹. However, the assumption of a constant lidar 153 154 ratio (extinction-backscatter ratio) throughout a profile causes a larger uncertainty in the 155 derivation of extinction when there is multi-layer aerosol transport [Welton et al., 2002].

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157 Level 2 quality-assured data products are currently under development, and beta level 158 2a aerosol data were made available for this study. Level 2a processing uses AERONET level 159 2 [Smirnov et al., 2000; Dubovik et al., 2002a; Holben et al., 2006] aerosol optical thickness, 160 and bad data are discarded in order to assure high quality. MPLNET data are discarded if one 161 or several of the following occurs: the data were acquired outside the preferred instrument 162 temperature range (22.5 ± 2 °C, for this instrument); the lidar ratio error was larger than 30%; 163 less than 80% of the signals in the 20-minute average were cloud free; or the signal-to-noise 164 ratio was higher than 20% directly above the top of the aerosol layer. MPLNET aerosol 165 products have been validated in a number of studies, and the most recent and comprehensive

one was by *Schmid et al.* [2006], which indicated the accuracy of beta level 2a MPLNET
aerosol extinction profiles to within 20%.

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169 **2.4. Data Usage**

170 The three years (from 2005 to 2007) of aerosol data (τ , α , ω , g, and σ) used in this 171 study were based on the AERONET level 2 and MPLNET beta level 2a data sets, which were 172 quality-assured and were only for cloud-free conditions. Figure 1a shows the number of days 173 in a month when the AERONET level 1 (not cloud-screened or quality-assured) and level 2 174 data were available at the two sites (EPA-NCU and NCU Taiwan). The AERONET data from 175 EPA-NCU were not available prior to 18 July 2006. Therefore, the data from the 176 NCU Taiwan site were used for this period. Three major issues affected the continuity of this 177 data set: (1) relatively few data were obtained from NCU Taiwan because this site did not 178 follow closely the AERONET standard measurement procedure for continuous monitoring 179 prior to 2009, (2) the Level 2 data from NCU Taiwan were eliminated due to an instrumental 180 issue (i.e., moisture contamination) during January and June 2006, and (3) a regular 181 calibration was performed for the EPA-NCU instrument during September and October 2007, 182 and there were no observations during these two months.

In Figure 1b, the MPLNET level 1 data set reveals the continuous measurements (day and night) with a total of 1034 days over 3 years (a data acquisition rate of 94 %). On the other hand, the MPLNET beta level 2a shows only 189 days over 3 years (a data acquisition rate of ~17%), because the MPLNET beta level 2a retrieval relies on AERONET level 2 data, which is cloud-screened and quality-assured data. In addition, if we consider a perfect measurement period (August 2006 to July 2007) when the MPL and the Sun/sky radiometer operated continuously, then the AERONET level 2 and MPLNET level 2a data for the
EPA-NCU site are 175 (~48%) and 119 (~33%) days per year, respectively. The value of 33%
represents the percentage of days with clear sky in a year and also the maximum available
days based on the measurement strategy for this site.

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194 Daily mean profiles were computed when at least three σ profiles were available in a 195 UTC day. There were a total of 145 daily mean σ profiles available at NCU for the analysis 196 of the vertical characteristics of the aerosol. The daily mean values of τ , α , ω , and g for the 197 145 days were calculated from

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$$\overline{x}_{\lambda} = \frac{1}{n} \cdot \sum_{i=1}^{n} x_{\lambda,i}$$
(1)

199 where $x_{\lambda,i}$ is the *i*th instantaneous observation for aerosol optical property *x* at the wavelength 200 λ , and *n* is the number of observations in a day. Due to a large variation in the days available 201 in each month, it is not proper to interpret the seasonal variation of the vertical distributions 202 of aerosol; and therefore, a classification for the vertical distribution of aerosol has been 203 applied in this study.

204

205 **3. Methodology**

206 **3.1. Vertical Distribution Classifications of Aerosols**

207 The σ profiles derived from the MPL observations were classified into three types in 208 order to describe the complex characteristics of aerosol vertical distributions. The rules for 209 the classification of these three types are listed in Table 1. Figure 2 shows the σ profiles of 210 the three types and the corresponding temperature and dew-point profiles. In Figures 2a–2c, 211 the daily mean σ profiles conform to the rules shown in Table 1, but the instantaneous σ 212 profiles show perturbations. The corresponding temperature and dew point temperature 213 profiles shown in Figures 2d-2f are the soundings at the Taipei station (~30 km north of 214 NCU). In Figures 2a and 2d (Type 1), the vertical distribution of mean σ is restricted to 215 within 2.0 km above ground level (AGL), which is known as the mixed layer [Stull, 1998]. 216 The sounding profiles show a strong and stable inversion layer around 2.0 km, which 217 confines aerosols to the region below 2.0 km. On the other hand, in Figure 2e, the sounding 218 profiles show an inversion layer around 1.7 km, which confines most of the aerosols in the 219 mixing layer (Figure 2b). In addition, a weaker mean σ was observed in the lower free 220 atmosphere (the layer of 1.7 - 2.8 km height in Figure 2e). This suggests that the source of 221 aerosol in the lower free atmosphere could be caused by three mechanisms: (1) the local 222 aerosols lift up to the free atmosphere from the mixing layer, (2) the aerosols remain in the 223 residual layer and (3) the long-range transport of the aerosols in the free atmosphere.

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225 Normally, mechanisms (1) and (2) can be identified based on the time evolution of 226 normalized relative backscatter (NRB) obtained from the MPLNET level 1 data. However, to 227 define the third mechanism, we need assistance from other information (such as back 228 trajectory, sounding and satellite data). For the Type 2 case (Figures 2b and 2e), the weaker 229 inversion layer of around 1.7 km and the well following ambient temperature and dew-point 230 profiles below 2.8 km imply the exchange of air mass between the mixing layer and the lower 231 free atmosphere, as evidenced by the NRB. However, in this case the contribution from the 232 long-range transport of aerosol in the free atmosphere is hard to separate. Compared to the 233 Figure 2b, Figure 2c (Type 3) shows enhanced σ between 2 and 4 km, which is most likely due to the long-range transport aerosol in the lower atmosphere. The strong inversion layer around 1.8 km, and the diverging ambient temperature and dew-point profiles between 2 and 3 km in Figure 2f provide evidence of the different sources of air mass in the vertical distribution. According to the characteristics of the vertical distributions of aerosol, in this paper Type 1 will refer to near-surface aerosol transport, Type 2 will refer to near-surface aerosol transport with upper-layer convective mixing/dispersion of aerosol, and Type 3 will refer to two-layer aerosol transport.

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3.2. Radiative Transfer Model Calculations

243 We investigated the aerosol radiative effect using the radiative transfer model of Chou 244 and Lee [1996] and Chou and Suarez [1999]. The model includes the absorption by ozone, 245 water vapor, oxygen and CO₂, as well as the absorption and scattering by clouds, aerosols, 246 and molecules (Rayleigh scattering). Fluxes are integrated virtually over solar spectrum, 247 ranging from 0.175 μ m to 10 μ m, which is divided into 11 bands in the model. Depending 248 upon the nature of the absorption, different approaches are applied to different absorbers. A 249 more detailed description of this model can be found elsewhere [cf. Chou and Suarez, 1999; 250 *Chou et al.*, 2006].

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Data input to this model includes the vertical profiles of temperature, humidity, ozone, surface albedo and the aerosol optical properties. In this study, we constructed 121 pressure layers in vertical, starting from the ground surface and ending up at the top-of-atmosphere (TOA), in which the layers of 1-81 (from surface to \sim 6 km AGL with the vertical resolution of 0.075 km) were based on the vertical profile of the MPL measurement. The temperature and humidity profiles taken from the 6-hourly NCEP reanalysis [*Kalnay et al.*, 1996] were interpolated to the model layers. The vertical ozone distribution was based on the climatology values. The daily surface albedo data [*Schaaf et al.*, 2002] were derived from the moderate resolution imaging spectroradiometer (MODIS) Collection 5 combined Level 3 16-day Albedo Products (MCD43B, http://ladsweb.nascom.nasa.gov/).

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263 The seven Cimel channels are located within the 11 bands of the radiative transfer model 264 of *Chou et al.* [2006]. We derived the aerosol optical properties of a model band by averaging 265 the AERONET-retrieved aerosol optical properties (i.e., τ , ω and g) within the band. For 266 those model bands outside the range of the Cimel channels, the aerosol optical properties 267 were set to be equal to those of the nearest Cimel channels [Chou et al., 2006]. Thus, the 268 vertical profiles of τ were derived by scaling the daily mean τ at each Cimel channel with the 269 MPLNET-retrieved σ vertical profiles at 527 nm. For the ω and g, we assumed constant 270 values in the vertical distribution. Because of the presence of clouds, aerosol retrievals may 271 not be complete in a day. It is therefore not feasible to derive the aerosol radiative effect using 272 diurnal variations of the aerosol optical properties. Instead, we used the daily mean values of 273 aerosol optical properties to derive hourly solar radiation, and then averaged to obtain the 274 corresponding daily mean solar radiation.

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The direct aerosol radiative effect (DARE, W m⁻²) is defined as the change in net radiation due to aerosols in clear-sky given by

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279
$$DARE(p) = F_{wa}(p) - F_{na}(p)$$
(2)

where F_{wa} (*p*) and F_{na} (*p*) represent the net downward fluxes with and without aerosols at pressure level *p*. Let DARE_{TOA} and DARE_{SFC} be the DARE at the TOA and the surface, respectively, the DARE in the atmosphere, DARE_{ATM}, which is the solar heating of the atmosphere due to aerosols, is then given by

$$DARE_{ATM} = DARE_{TOA} - DARE_{SFC}$$
(3)

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Finally, the impact of aerosols on the atmospheric heating rate of a layer between *p* and $p+\Delta p$, 289 $\Delta Q(p)$, is proportional to $DARE(p) - DARE(p+\Delta p)$.

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291 4. Results and Discussion

292 4.1. Vertical Distribution of Aerosols

293 Three types of σ profiles were classified based on the conditions described in section 294 3.1. These 3 types were found in 28, 51, and 31 days, respectively, and comprised 76% of the 295 145 days. The vertical profiles of the mean and standard deviation of the σ of Types 1–3 are 296 shown in Figures 3a–3c, respectively. Below 337.5 m AGL, no readings of the σ are shown 297 due to the near-field observation limits of our particular MPL [Campbell et al., 2002]. 298 Specifically, the average calculation for the daily σ profiles not only smoothes the profiles, 299 but also eliminates some peak values. In addition, the large standard deviation shows that the 300 classification we applied in this study is an approximation. The following sections provide a 301 brief discussion of the σ profiles type classifications in relation to the backward trajectories 302 for each type.

304 *Type1: Near-surface aerosol transport*

In Figure 3a, the mean σ decreases with the increase in height, and is restricted within 2.0 km. The highest σ shows ~0.2 km⁻¹ near 0.5 km and decreases to ~0.01 km⁻¹ at ~2.0 km. The height of 2 km represents the maximum mixing layer height for Type 1, in which the mixing layer height varies between 1.0 km and 2.0 km as illustrated by the standard deviations of profile. In addition, the fall and winter seasons (September - January) are prevailing seasons for the Type 1 classifications (Figure 3d), but few days were observed in the summer time.

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313 Trajectory analysis has been used to diagnose the movement of pollutants in previous 314 studies [e.g., Draxler, 1996; Wang et al., 2007 and references therein]. The Hybrid 315 Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model, developed by NOAA 316 Air Resources Laboratory [Draxler and Rolph, 2003], is applied to calculate air mass back 317 trajectories. The meteorological data used to initialize HYSPLIT is obtained from the NCEP 318 Global Data Assimilation System (GDAS) data set [Kalnay et al., 1996]. Figure 4 shows the 319 5-day backward trajectories of Types 1–3 ending at 0.5 km and 2.5 km above the mean sea 320 level (MSL) over NCU. The altitudes of 0.5 km and 2.5 km present the near-surface and 321 upper-layer air mass transport, respectively. The color table uses in the image denotes 322 trajectory altitudes for the 5-day computational period. The symbols mark the days behind the 323 ending day.

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In Type 1 (Figure 4a), the major pathway of the air mass shows the potential aerosol

326 source regions from northeast Asia with trajectories passing over the West Pacific. The four 327 trajectories that strayed from the major pathway correspond to the days in spring and summer 328 as shown in Figure 3d. Most of the trajectories traversed over coastal regions which have 329 been identified as major anthropogenic regions in East Asia [Street et al., 2003]. When the air 330 mass traversed over the ocean during the last two days, the trajectories were found to be 331 mainly within the marine boundary layer (below 1.0 km in the color table). The near surface 332 shallow transport with anthropogenic aerosol (e.g., sulfate and nitrate) over the ocean is 333 expected to have more aerosol water uptake or more aerosol hydroscopic growth [Seinfeld, 334 1986]. In the final half day before ending at NCU, the trajectories pass over northern Taiwan 335 where the local pollutants also contribute to the aerosol loading. In general, the Type 1 336 classifications are represented by the northerly flow causing a transport belt for air mass 337 advection over long distances towards Taiwan. The σ below 2 km are due to the presence of 338 an abundance of anthropogenic aerosols caused by long-range transport and local pollutants.

339

340 Type 2: Near-surface aerosol transport with upper-layer convective mixing/dispersion of
341 aerosols

In Figure 3b, the mean σ decreases with the increase in height, and is restricted to within 4.0 km. The two σ peaks below 1.0 km with a value of ~0.27 km⁻¹ illustrated the various maximum σ heights in Type 2 cases. Compared to Type 1, the mean σ profile of Type 2 shows an aerosol layer that is extended up to 4 km. The upper-layer aerosol transport related to the convective mixing/dispersion of aerosols in the free atmosphere is described in Section 3.1. Figure 3e shows the diverse coverage for months in Type 2, in which the most frequent occurred month is July.

350 The 5-day backward trajectories at 0.5 km of NCU for Type 2 are shown in Figure 4b. 351 Most of the trajectories come from the north of Taiwan via a route similar as that of Type 1. 352 This implies that similar atmospheric features may represent the near-surface aerosol 353 transport observed in both Types 1 and 2. In addition, some of the southern and eastern 354 trajectories related to the summer season also belong to Type 2. These trajectories originate 355 from the clean marine atmosphere and from the atmosphere in Southeast Asia where low 356 pollution emissions are found during the summer. As a result, local emissions might be the 357 essential source of the near-surface aerosol layer. For the upper-layer transport, as shown in 358 Figure 4d, the trajectories ending at 2.5 km show various routes and are mostly westerly in 359 winter and southeasterly in summer. These westerly trajectories will have traveled over densely populated and industrialized areas. In these regions the air motion transports the 360 361 aerosols to the free atmosphere by means of frontal lifting [e.g., Bey et al., 2001; Liang et al., 362 2004]. On the other hand, the southeasterly trajectories which originated above the West 363 Pacific Ocean, pass over the Philippines, the South China Sea, off the coast of southern China, 364 and then over southern Taiwan. Most likely the marine pathway has less long-range transport 365 aerosols from the continental regions, but a higher possibility of local aerosol aloft due to the 366 enhanced vertical convection mixing of aerosols in summer. However, the aerosol particles 367 that lift towards the free atmosphere are not able to be seen in the HYSPLIT simulation 368 because of the coarse resolution of the meteorological data, as well as the terrain data.

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370 *Type 3: Two-layer aerosol transport*

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The two-layer transport structure of aerosols has been reported in the literature [e.g.,

372 Welton et al., 2002; Murayama et al., 2004; He et al., 2008; Johnson et al., 2008; McFarlane 373 et al., 2009]. An MPL retrieved mean σ profile with a 2-layer structure (Type 3) over NCU is 374 shown in Figure 3c. In contrast to Type 2, Type 3 shows significant upper-layer (2–4 km) aerosol transport. The prominent σ of ~0.09 km⁻¹ at 2.4 km reveals almost one-half the 375 376 amount of ~ 0.21 km⁻¹ near the ground, thereby emphasizing the importance of upper-layer 377 transport. We suggest that the 2-layer aerosol transport is not only due to considerable 378 quantities of long-range transport aerosols exist in the free atmosphere but also the fact that 379 inversion layer plays a significant role in preventing the mixing of vertical convective aerosol. 380 Figure 3f shows that except for the unusual occurrence of a frequency of 10 days in October 381 which related to a single event during 11 - 23 October 2006, the most frequent occurrence for 382 2-layer aerosol transport season take place in spring (March – May), which agrees with 383 previous studies in this region [Murayama et al., 2004; Chiang et al., 2007].

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385 The trajectory distribution of Figure 4c shows a distribution similar to that of Figure 4b, 386 which implies that the near-surface aerosol transport has similar aerosol characteristics for 387 Type 2 and Type 3. In terms of backward trajectories at 2.5 km in Type 3 (Figure 4e), the 388 westerly trajectories suggest that westerlies prevail in the middle troposphere over 389 mid-latitude East Asia. Most of the trajectories originate in Indochina and travel slowly over 390 Southeast China during the last three days. In their pathway, air masses are likely to carry 391 biomass-burning aerosols from Indochina and mix them with the abundance of anthropogenic 392 aerosols from Southeast China and then advect them downwind towards northern Taiwan in 393 the lower free atmosphere. The spatial distribution of τ from the MODIS measurement will 394 be further applied to understand the sources region of aerosol in the next subsection.

396 4.2. Spatial Distribution of Aerosol Optical Thickness

397 The MODIS-Aqua Level 3 data with 1°×1° resolution were obtained from the Giovanni 398 online data system (http://disc.sci.gsfc.nasa.gov/giovanni/) in order to analyze the spatial 399 distribution of aerosols in relation to each classified type. MODIS sensors perform near 400 global daily observations of aerosols and eight of the 36 channels (between 0.412 and 2.13 401 μm) are used to retrieve aerosol properties over land [Kaufman et al., 1997b; Hsu et al., 2004 402 and 2006; Levy et al., 2007] and ocean [Tanré et al., 1997]. Figure 5 shows that the 403 MODIS-Aqua Level 3 aerosol retrievals of averaged τ at 550 nm for Types 1–3 in the Asian 404 region. Based on the trajectory analysis in previous section, a lead-time of three days was 405 selected for MODIS-retrieved τ to compute the average for each Type of aerosol transport.

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407 The main locations of the large τ in Figures 5a–5c (coastal regions of China, central 408 China, and the Indo-Gangetic Plain) correspond with the relatively densely populated and 409 industrial areas. The potential aerosol sources at NCU for each type can be further illustrated 410 by combining them with the back trajectory in Figure 4. In Figure 5a, the high τ (~1) over the 411 northeastern coast of China (near 35°N, 118°E) have a high probability of contributing to the 412 near-surface aerosol transport in Type 1. On the other hand, it is evident from the Figures 5b 413 and 5c that τ are higher than those over the Asian continent and with a stronger continental 414 outflow over the Yellow Sea (near 36°N, 123°E). According to the suggestion of trajectory 415 analysis, the air masses of the near-surface aerosol transport of Type 2 traversing over the 416 high τ region in northeastern China appear more frequently indicating a higher mean σ of 417 near-surface aerosol transport in Type 2 (Figure 3b). On the other hand, the high τ over southeastern China conform to the potential pollution source regions of the upper-layer aerosol transport in Type 3. The high τ over southeastern China are related to the local emission sources [*Street et al.*, 2003] and the transported smoke aerosols from the biomass burning in Southeast Asia in the spring [*Hsu et al.*, 2003; *Wang et al.*, 2007].

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423 4.3. Aerosol Optical Thickness and Ångström Exponent

Table 2 shows the average and standard deviations of aerosol optical properties for Types 1–3. The mean values of τ_{500nm} are 0.28, 0.45, and 0.44 for Types 1–3, respectively, and are related to the integrated σ profiles. On the other hand, the $\alpha_{440-870nm}$ remains nearly constant with a mean of ~1.4 for these 3 types, implying that fine mode particles dominate the pollutants. Fine mode particles are commonly observed in the long-range transport of anthropogenic aerosols [*Eck et al.*, 2005].

430

431 Figure 6 presents the scatter plots of the AERONET-retrieved daily mean $\alpha_{440-870nm}$ vs. 432 τ_{500nm} for Types 1–3. The error bars present a one standard deviation. Basically, most data 433 have $\alpha_{440-870nm}$ values within the range of 1.0–1.8, except for 2 days with lower $\alpha_{440-870nm}$ (< 434 1.0) of Type 2, implying that the common feature of fine mode aerosols dominate the 435 pollutants for those 3 classified types. However, the two days with low $\alpha_{440-870nm}$ reflect the 436 occurrence of coarse mode particles in Type 2. In the case of lowest $\alpha_{440-870nm}$ (~0.8) on 5 437 July 2007 in Figure 6b, the aerosols may possibly have been contaminated by cirrus clouds 438 [Smirnov et al., 2000]. The other low $\alpha_{440-870nm}$ case in Figure 6b happened on 28 January 439 2007, when an Asian dust storm was reported by Taiwan EPA. In terms of aerosol loading, 440 the ranges of τ_{500nm} show 0.13–0.63, 0.15–1.05, and 0.13–1.17 for Types 1–3, respectively.

441 Similar minimum values of τ_{500nm} suggest that those 3 classified aerosol profiles can be 442 observed even in a low aerosol loading atmosphere. On the other hand, the cases with τ_{500nm} 443 higher than 0.6 are only found in Types 2 and 3. Those high τ_{500nm} cases are not only caused 444 by the upper-layer aerosol transport but also by the stronger aerosol emission sources shown 445 in Figure 5. Consequently, we suggest that a fairly near-surface aerosol transport can 446 contribute to the daily mean τ_{500nm} reaching as high as 0.6. However, when a daily mean 447 τ_{500nm} greater than 0.6 is observed, it implies that an upper-layer aerosol transport could occur, 448 and that the aerosol vertical distribution should be further considered for estimating the 449 radiative effect of aerosol.

450

451 4.4. Single-scattering Albedo and Asymmetry Factor

452 The estimated DARE of the Earth-atmosphere system has to rely on data sources for ω 453 and g, and especially the data on ω are particularly critical for determining the partition of the 454 DARE between the atmosphere and the underlying surface [Chou et al., 2006]. By definition, 455 ω is obtained by dividing the aerosol scattering coefficient by the aerosol extinction 456 coefficient, and g is the fraction of the incident radiation scattered forward after striking an 457 aerosol. g = 1 if 100% of the incident radiation is scattered forward, whereas the g = 0458 indicates that one half of the incident radiation is forward-scattered while the other half is 459 backscattered. The spectral ω and g for Types 1–3 are plotted in Figure 7. Valid data have 460 been selected by the AERONET level 2 inversion data described in section 2.2, and the 461 numbers of the daily mean for Types 1–3 are 4, 24, and 10 days for ω , and 20, 38, and 19 462 days for g. Group-mean spectral ω and g are derived by averaging arithmetically all the 463 available daily-mean values of ω and g of a given type. The group-mean and one standard

464 deviation of ω and g at 440 nm for Types 1–3 are listed in Table 2.

465

In Figures 7a–7c, most of the daily mean ω ranging from 0.90 to 0.98 in any wavelength shows that the aerosols are moderately absorbing. In comparison, *Eck et al.* [2005] indicated that in Beijing (China) and Anmyon Island (Korea) the ranges of ω were within 0.84–0.92 and 0.88–0.95, respectively, suggesting that the aerosols had a stronger scattering property at NCU. The stronger scattering property (higher ω) could be caused by hygroscopic growth but also with other possible aging mechanisms such as gas-to-particle conversion, condensation, and coagulation [*Eck et al.*, 2005].

473

474 The characteristic of the ω changing with the increase in wavelength relates to the 475 aerosol types for each individual day as shown in Figures 7a–7c. The relationships between 476 spectral dependence of ω and the key aerosol types (e.g., urban-industrial, biomass-burning, 477 desert dust and marine aerosols) have been described in *Dubovik et al.* [2002a], and are based 478 on eight years of worldwide distributed data from the AERONET network. In Type 1, the 479 characteristics of spectral dependence of ω exhibit two different types of aerosol (Figure 7a). 480 Two days (30 - 31 January 2007) with the ω increasing with the increase in wavelength 481 represented the characteristic of desert dust aerosols [Dubovik et al., 2002a; Eck et al., 2005], 482 and has also been identified as a dust event by the Taiwan EPA announcement. On the other 483 hand, the other two days with the ω decreasing with the increase in wavelength show that the 484 characteristic of the urban-industrial aerosol [Dubovik et al., 2002a], and the values of ω 485 show comparable ranges to those of Anmyon Island in Korea [*Eck et al.*, 2005]. For Type 2, 486 most days presented the urban-industrial aerosol type, but a few days presented the desert 487 dust aerosol type. A case with an extreme low ω with the characteristic of the ω decreasing 488 with the increase in wavelength was observed on 29 March 2007, which was linked to the 489 biomass-burning aerosols type described in *Dubovik et al.* [2002a]. Compared to Types 1 and 490 2, Type 3 (Figure 7c) shows lower spectral dependence of ω with higher ω values ranging 491 from ~ 0.92 to 0.97. This implies that the aerosols in the upper-layer likely have a stronger 492 scattering property. Overall, the spectral dependence of ω shows that the downwind area of 493 Asian continental pollutants is dominated by urban-industrial aerosols of three types. 494 However, the observed desert dust, the biomass-burning and the mixing aerosols emphasize 495 the complex aerosol types in this region.

496

497 In Figures 7d–7f, most of the daily mean g ranged from 0.60 to 0.75 in any wavelength 498 inhibit the aerosols tend to scatter more energy forward. The spectral dependence of g 499 decreases with the increase in wavelength, illustrating that the aerosols have more forward 500 scattering in short wavelengths. As to the group-mean spectral g, the slope of g with 501 wavelength in Type 1 is lower than in Types 2 and 3. The lower slope of the spectral g with a 502 higher g_{1020} in Type 1 represents the characteristic of urban-industrial, desert dust and marine 503 aerosols [Dubovik et al., 2002a]. On the other hand, a case (1 April 2007) of Type 3 shows a 504 pronounced decrease of the g to the relatively low values (from $g_{440} = 0.71$ to $g_{1020} = 0.52$) 505 which corresponds to the features of biomass-burning aerosols [Dubovik et al., 2002a].

506

507

4.5. Aerosol Radiative Effect and Heating Rate

508 The scatter plots of the daily mean DARE_{TOA} and DARE_{SFC} vs. τ at 500 nm are shown 509 in Figure 8. Straight lines indicate the linear regressions. Each point represents one day.

510 Circles represent radiative calculations on those days with daily-mean spectral τ , ω and g. 511 Crosses represent calculations on those days where there are only daily-mean spectral τ , but 512 no daily-mean spectral ω and g. Group-mean spectral ω and g are applied in the calculations. 513 Squares represent calculations on those days where there are daily-mean spectral τ and g, but 514 no daily-mean spectral ω . The group-mean spectral ω are applied in the calculations. The DARE is highly linearly correlated with τ_{500nm} with a negative correlation coefficient of > 515 516 0.93 for all types of aerosol vertical distributions, indicating that DARE can be reliably 517 estimated from column-integrated τ_{500nm} . However, some points show a significant deviation 518 from the linear regression which is related to the broad ranges of observed ω and g, especially 519 the DARE_{SFC} in Type 3. The large negative deviation of the DARE_{SFC} from the linear 520 regression on 1 April 2007 (Figure 8f) is due to the small ω and g (Figures 7c and 7f). On the 521 other hand, the case of 14 October 2006 with large ω and g shows a weak negative DARE_{SEC}. 522 The deviation of DARE from the linear regression can reach up to ± 10 W m⁻².

523

At the TOA, the mean values of DARE_{TOA} are -7.1, -11.6, and -11.2 W m⁻² for Types 1, 2, 524 525 and 3, respectively. The negative DARE_{TOA} implies that the effect of the reflection of solar 526 radiation due to aerosols is larger than the effect of absorption, and that the net effect is a 527 cooling of the earth-atmosphere system. At the surface, the mean values of DARE_{SFC} are -13.2, -20.3, and -18.7 W m⁻² for Types 1, 2, and 3. The significant decrease of the solar radiation at 528 529 the surface is enhanced by the absorption of solar radiation in the atmosphere due to aerosols. 530 The absorption of solar radiation due to aerosols in the atmosphere, DARE_{ATM}, is 6.1, 8.7, and 7.5 W m⁻² for Types 1–3, respectively. 531

533 The aerosol radiative efficiency (ARE) is defined as the change of DARE per unit 534 change of τ at 500 nm. This parameter is useful for quantifying and comparing aerosol 535 radiative effects at different places under a wide range of aerosol conditions [Wang et al., 536 2007 and references therein]. We estimated ARE_{TOA} and ARE_{SFC} from the slopes of the linear 537 regressions shown in Figure 8, in which uncertainties can be explained by root-mean-square 538 (rms). The AREs for Types 1–3 are listed in Table 3. Values of ARE_{TOA} for these three types are very close (~-23 W m⁻²), indicating that the solar energy budget of the earth-atmosphere 539 540 system is not overly sensitive to the vertical distributions of aerosols over northern Taiwan. 541 Compared to ARE_{TOA}, the ARE_{SFC} and ARE_{ATM} show relative sensitivity to the vertical distributions of aerosols, with the highest efficiency in Type 1 and the lowest efficiency in 542 543 Type 3.

544

545 A sensitivity study was performed to investigate how the aerosol vertical distributions 546 shaping the computations of ARE under the assumption of unified aerosol optical properties 547 (i.e., fixed column-integrated τ , column-mean ω , and g of moderate-absorbing aerosol) for 548 various profiles. As expected, the results show that all ARE_{TOA}, ARE_{SFC} and ARE_{ATM} increase 549 (i.e., larger negative values for the first two but larger positive value for the last ARE) when 550 more aerosols are elevated to higher levels in various profiles within thin- τ regime. These 551 increases are due to more downwelling solar irradiance available for aerosols to interact at 552 higher levels, leading to enhanced reflection at TOA and absorption in ATM (in turn, dimmed 553 transmission at SFC). However, as the degrees of freedom in aerosol properties increase (e.g., 554 ω , g, τ , in addition to vertical distribution) in model simulations, the variation of AREs 555 becomes complex due to their competing nature for solar irradiance. Since most of aerosols in Types 1–3 are confined in the lower atmosphere (i.e., below 3 km) with moderate absorption, the values of ARE_{TOA} shown in Table 3 are weakly sensitive to aerosol vertical distributions. When the variations of other aerosol properties (*cf.* Figures 3 and 7) come into play (e.g., ω_{λ} dominant in Type–1 *vs.* Type–2, against their similar vertical distributions; or larger variability of ω_{λ} and vertical distribution in Type–3 *vs.* Type–2), ARE_{SFC} coupled with ARE_{ATM} reveal relatively larger variability, compared to ARE_{TOA}.

562

563 Although we have demonstrated that the vertical distribution of aerosols does not make 564 a significant difference in the estimation of ARE_{TOA}, it does provide information as to the 565 impact of aerosols on the vertical profile of the atmospheric heating rate. Figure 9 shows the vertical distribution of the mean and one standard deviation of ΔQ (K day⁻¹) for Types 1–3. The 566 567 ΔQ profiles are similar to the σ profiles in Figure 3, with maximum values of 0.37, 0.41, and 0.26 K day⁻¹ at around 700 m in height for Types 1–3, respectively. A larger σ implies a higher 568 569 aerosol concentration, and hence a stronger impact on ΔO . The ΔO of Type 3 is smaller than 570 those of Types 1 and 2 below 2 km, indicating that the upper-layer absorbing aerosol transport 571 enhances the upper-layer ΔQ at the expense of reducing the near-surface ΔQ . The nearly 572 constant ΔQ in the vertical implies a weaker impact on the stability and convection. On the 573 other hand, Types 1 and 2 have a larger ΔQ near the surface and, hence, have a larger impact on 574 convection than Type 3.

575

576 4.6. Improving ARE Estimates for Two-layer Aerosol Transport

577

The assumption of a constant column-mean ω and g throughout a two-layer aerosol

578 transport profile may cause an error in the radiative transfer calculations. The coexistence of 579 near-surface and upper-layer aerosol transport as shown from different source regions, 580 implies different ω and g in the vertical distribution. For example, the different ω in the 581 near-surface and upper layers has been observed using airborne measurements over Niamey 582 [Osborne et al., 2008]. Except for aircraft measurement, the currently available retrieval 583 schemes based on ground measurements are unable to resolve vertical variations of ω and g. 584 Nevertheless, the AERONET measurements at a high mountain site at Lulin (23.51°N, 585 120.92°E; 2862 m MSL; ~180 km south of NCU), can provide useful information on the 586 optical properties of aerosols for the upper layers of Type 3 at NCU under the assumptions of 587 similar backward trajectories and aerosol sources [*Wai et al.*, 2008, Figure 3].

588

589 The averaged ω and g of Lulin obtained from AERONET were used as surrogate to 590 improve the ARE estimates for the 2-layer aerosol transport. Due to the quality control 591 limitations of the AERONET inversion algorithm, only a few days of level 2 data [Holben et 592 al., 2006] are available for 2007–2008. They occurred in March–May except for one day in 593 August. The available level 2 data give a 4-day averaged ω_{440nm} of 0.96 and a 16-day 594 averaged g_{440nm} of 0.7. Aerosols in the upper layer observed at Lilun exhibited a relatively 595 stronger scattering property and backward scattering, compared to Types 1–3. Here, we apply 596 aerosol optical properties derived from the Sun/sky radiometer at Lulin, to the upper-layer 597 aerosol transport in Type 3 and repeat the simulations of aerosol effect. In this case, the 598 process basically follows the vertical structure of Type 3, but the ω and g for the near-surface 599 aerosol layer were replaced with group-average values of Type 1 and 2 and for upper-layer 600 were replaced with averaged values of Lulin. As a result, the change in the vertical

601 distribution of the ω and g affects on the ARE calculations are shown in Table 3. Compared 602 to the results of Type 3 in Table 3, the redistributions of ω and g in the vertical profiles enhance the negative ARE_{SFC} (surface cooling, \sim -3 W m⁻²) and the positive ARE_{ATM} 603 (atmosphere heating, $\sim 3 \text{ W m}^{-2}$), but have only a minor influence on ARE_{TOA}. In addition, the 604 vertical distribution of ΔQ shows an enhanced ΔQ by up to 0.2 K day⁻¹ in the surface layer 605 606 when near-surface aerosol replaced with the group-mean aerosol ω and g of Types 1 and 2 607 (shown in Figure 9c). These preliminary results encourage us to approach the 2-layer aerosol 608 transport problems in the future with case studies using the data from EPA-NCU and the Lulin 609 sites.

610

611 5. Conclusions

In this study, we presented a synergistic process to determine the vertical distributions of aerosol optical properties and the impact of aerosols on solar heating using ground-based remote-sensing (Micro-pulse lidar and Sun/sky radiometers) and a radiative transfer model. The MPL and Sun/sky radiometer data were taken from observations at a rural site in northern Taiwan and covered a period of three years (2005–2007).

617

To simplify the complex characteristics of the aerosol vertical distribution, lidar-retrieved aerosol extinction profiles were classified into three types with common characteristics of near-surface aerosol transport. These three types of aerosol profiles account for 76% of the total data base. In general, the common near-surface aerosol transport (0 to 2 km) is related to the northerly air mass with possible long-range transport of aerosols during wintertime. The upper-layer aerosol transport (2 to 4 km) is caused by the convective

624 mixing/dispersion of aerosol in the free atmosphere.

625

626 The columnar aerosol optical properties show that the aerosols in this region are 627 dominated by fine mode (Ångström exponent = ~ 1.40) and moderately absorbing aerosols (ω = ~ 0.93 and g = ~ 0.73 at 440 nm), which are typical of urban-industrial aerosols [Dubovik et 628 629 al., 2002a]. However, frequent perturbations due to desert dust, biomass-burning or mixed 630 aerosols suggest a diversity of aerosol types over the downwind area of the Asian continent. 631 Compared to the ω in the upwind areas (China and Korea), the higher ω in northern Taiwan 632 suggests that anthropogenic aerosols transported near the ocean surface are likely to 633 experience hydroscopic growth. The column-integrated aerosol optical thickness at 500 nm 634 (τ_{500nm}) ranges from 0.1 to 0.6 for near-surface aerosol transport, but can be doubled in the 635 presence of upper-layer aerosol transport.

636

637 Absorbing aerosols have the effect of warming the atmosphere and cooling the surface. The sensitivity of solar radiation to a unit change of τ_{500nm} , referred to aerosol radiative 638 639 efficiency (ARE), was computed at the top of the atmosphere (ARE_{TOA}) and at the surface 640 (ARE_{SEC}). The ARE_{TOA} is not sensitive to the vertical distributions of aerosols, and is approximately -23 W m⁻² for the three types of aerosol profiles. On the other hand, the 641 642 ARS_{SFC} is relatively sensitive to the vertical distribution of aerosols, and is -44.3, -40.6 and -39.7 W m⁻² for the near-surface, mixed and two-layer transport types, respectively. The 643 644 difference is caused primarily by the difference in ω ; surface transport type aerosols have the 645 smallest ω , and the two-layer transport type aerosols have the largest ω . Correspondingly, the 646 impact of aerosols on the vertical profile of solar heating is the largest for the near-surface transport type and the smallest for the two-layer transport type. Since any changes of solar heating in the atmosphere and at the surface affect the stability of the atmosphere, different aerosol transport type will have different impact on the atmospheric stability, convection, and regional climate.

651

652 The coexistence of near-surface and upper-layer aerosol transports infers different 653 aerosol characteristics in the vertical distribution. This study introduce a method, using 654 Sun/sky radiometer observation at the high-altitude station, Lulin (2862 m), to further assist 655 in estimating the upper-layer aerosol optical properties (ω and g) and improve the ARE 656 calculations for the two-layer transport type of aerosols. Using the proposed method, the recalculated ARE_{SEC} and ARE_{ATM} are enhanced by 3 W m⁻², and the solar heating is enhanced 657 by up to 0.2 K day⁻¹ in the surface layers. This analysis shows the sensitivity of the vertical 658 659 distribution of aerosol optical properties for estimating the aerosol radiative effect.

660

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821 Figure captions

Figure 1. Histogram of the number of days in the month for: (a) AERONET and (b) MPLNET data sets. The light gray and dark gray color bars present level 1 and level 2 data, respectively, for both of the AERONET and the MPLNET data sets. The black color bar presents the MPLNET level 2 dataset with data from Types 1–3 only.

Figure 2. Examples of aerosol extinction (km⁻¹) profiles obtained by MPL at EPA-NCU for: 826 827 (a) Type 1 (12-Nov-2006), (b) Type 2 (13-Feb-2007) and (c) Type 3 (15-Oct-2006). The blue 828 line is the daily-averaged profile and the red lines present all profiles on that day. Below 829 375.0 m, no readings are shown due to the near-field observation limits of MPL. Examples of 830 the ambient (Square point) and dew point (dot point) temperature (°C) profiles obtained by 831 Taipei sounding station (~ 30 km north of NCU) at 00UTC for: (d) Type 1 (12-Nov-2006), (e) 832 Type 2 (13-Feb-2007) and (f) Type 3 (15-Oct-2006). The dashed lines between two points 833 present continuously sampling in the vertical distribution, otherwise invalid values are noted.

- Figure 3. Profiles of the mean aerosol extinction with one standard deviation derived from the
- 835 MPLNET observations at EPA-NCU for (a) Type 1, (b) Type 2 and (c) Type 3, with
- corresponding monthly total number of days for (d) Type 1, (e) Type 2 and (f) Type 3.

Figure 4. Five-day backward trajectories of NCU for (a) Type 1 starting at 500 m, (b) Type 2 starting at 500 m, (c) Type 3 starting at 500 m, (d) Type 2 starting at 2500 m, and (e) Type 3 starting at 2500 m. Each trajectory starts at 00 UTC. Trajectory altitudes (based on the mean sea level, in meters) are denoted by the color scale.

Figure 5. Composite MODIS-Aqua aerosol Deep-Blue and Dark-Target retrievals of τ_{550nm}

- averaged for (a) Type 1, (b) Type 2, and (c) Type 3. The third day before the case days based
- on the potential pollution source regions has been applied in these plots.
- Figure 6. Scatterplots of τ_{500nm} vs. $\alpha_{440-870nm}$ at NCU for (a) Type 1, (b) Type 2 and (c) Type 3.
- 845 The error bars indicate the one standard deviation.
- Figure 7. The AERONET inversion data (ω and g) at 440, 675, 870, 1020 nm at NCU for (a)
- 847 ω of Type 1, (b) ω of Type 2, (c) ω of Type 3, (d) g of Type 1, (e) g of Type 2, and (f) g of
- 848 Type 3. The daily-mean spectral ω and g are shown as cross points and thin lines. The 849 group-mean spectral ω and g are shown as black circles and bold lines.
- Figure 8. Scatterplots of DARE at TOA vs. τ at 500 nm for (a) Type 1, (b) Type 2, and (c) Type 3. Scatterplots of DARE at the surface vs. τ_{500nm} for (d) Type 1, (e) Type 2, and (f) Type 3. Circles represent radiative calculations based on daily-mean spectral ω and g, crosses represent data based on only group-mean spectral ω and g, and squares represent data based on daily-mean spectral g and group-mean spectral ω .
- Figure 9. The vertical distribution of the mean and one standard deviation of the impact of
- aerosols on the atmospheric heating rate ΔQ (K day⁻¹) over NCU for (a) Type 1, (b) Type 2
- and (c) Type 3 (red lines). The green line* and the blue line** show the improved ΔQ profiles
- for Type 3. See Table 3 for detailed descriptions of * and **.

Type 1	1. The column-integrated τ_{527nm} greater than 0.1
	2. No σ are detected above 6 km AGL [*]
	3. A remarkable and isolated near surface aerosol layer
Type 2	1. Same as rules 1–3 in Type 1
	2. Above the near surface aerosol layer, presenting a weaker aerosol
	layer with vertical aerosol mixing
Type 3	1. Same as rules 1–3 in Type 1
	2. Above the near surface aerosol layer, presenting another remarkable
	and isolated aerosol layer
	3. The joint between the two layers shows low σ

859 Table 1. The classification rules for daily mean aerosol extinction (σ) profiles

*According to long-term MPL observations of EPA-NCU, most atmospheric aerosols exist
below 6 km AGL at this site.

862

864 Table 2. The aerosol optical thickness (τ) at 500 nm, Ångström exponent (α) at 440 - 870 nm, 865 single-scattering albedo (ω) at 440 nm, and asymmetry factor (g) at 440 nm retrieved from 866 Sun/sky radiometer measurements

Туре	Days	τ_{500nm}		α _{440–870nm}		<i>w</i> _{440nm}		<i>B</i> 440nm	
		mean	std	mean	std	mean	std	mean	std
Type 1	28	0.28	0.14	1.40	0.17	0.92	0.02	0.72	0.02
Type 2	51	0.45	0.22	1.40	0.18	0.95	0.02	0.73	0.02
Type 3	31	0.44	0.26	1.43	0.15	0.95	0.02	0.73	0.02

	ARE _{TOA}	ARE _{ATM}	ARE _{SFC}
Type 1	-23.3	+21.0	-44.3
Type 2	-22.6	+18.0	-40.6
Type 3	-22.9	+16.8	-39.7
Type 3 (Lulin1*)	-23.1	+19.8	-42.9
Type 3 (Lulin2**)	-24.2	+16.9	-41.1
			Unit: W m ⁻²

868 Table 3. Aerosol Radiative Efficiency (ARE) of TOA, ATM and SFC for Types 1–3.

* Values of ω and g at the upper-layer and the near-surface layer of Type 3 were replaced with the mean values from the Lulin site and Type 1, respectively.

872 ^{**} Values of ω and g at the upper-layer and the near-surface layer of Type 3 were replaced

873 with the mean values from the Lulin site and Type 2, respectively.





878 level 2 dataset with data from Types 1–3 only.



Figure 2. Examples of aerosol extinction (km⁻¹) profiles obtained by MPL at EPA-NCU for: (a) Type 1 (12-Nov-2006), (b) Type 2 (13-Feb-2007) and (c) Type 3 (15-Oct-2006). The blue line is the daily-averaged profile and the red lines present all profiles on that day. Below 375.0 m, no readings are shown due to the near-field observation limits of MPL. Examples of the ambient (Square point) and dew point (dot point) temperature (°C) profiles obtained by Taipei sounding station (~ 30 km north of NCU) at 00UTC for: (d) Type 1 (12-Nov-2006), (e) Type 2 (13-Feb-2007) and (f) Type 3 (15-Oct-2006). The dashed lines between two points present continuously sampling in the vertical distribution, otherwise invalid values are noted.



888 Figure 3. Profiles of the mean aerosol extinction with one standard deviation derived from the MPLNET observations at EPA-NCU for (a) Type

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890 Sole 100E 110E 120E 130E 140E Sole 100E 110E 120E 130E 140E
891 Figure 4. Five-day backward trajectories of NCU for (a) Type 1 starting at 500 m, (b) Type 2 starting at 500 m, (c) Type 3 starting at 500 m, (d)
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Figure 5. Composite MODIS-Aqua aerosol Deep-Blue and Dark-Target retrievals of τ_{550nm} averaged for (a) Type 1, (b) Type 2, and (c) Type 3.

897 The third day before the case days based on the potential pollution source regions has been applied in these plots.



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902

Figure 7. The AERONET inversion data (ω and g) at 440, 675, 870, 1020 nm at NCU for (a) ω of Type 1, (b) ω of Type 2, (c) ω of Type 3, (d) gof Type 1, (e) g of Type 2, and (f) g of Type 3. The daily-mean spectral ω and g are shown as cross points and thin lines. The group-mean spectral ω and g are shown as black circles and bold lines.



Figure 8. Scatterplots of DARE at TOA vs. τ at 500 nm for (a) Type 1, (b) Type 2, and (c) Type 3. Scatterplots of DARE at the surface vs. τ_{500nm} for (d) Type 1, (e) Type 2, and (f) Type 3. Circles represent radiative calculations based on daily-mean spectral ω and g, crosses represent data based on only group-mean spectral ω and g, and squares represent data based on daily-mean spectral g and group-mean spectral ω .



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Figure 9. The vertical distribution of the mean and one standard deviation of the impact of aerosols on the atmospheric heating rate ΔQ (K day⁻¹) over NCU for (a) Type 1, (b) Type 2 and (c) Type 3 (red lines). The green line* and the blue line** show the improved ΔQ profiles for Type 3. See Table 3 for detailed descriptions of * and **.