

1 **Aerosol optical characteristics in the urban area of Rome, Italy, and their impact**
2 **on the UV index.**

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17

18 **Abstract**

19 The impact of the aerosol optical properties on the ultraviolet index (UVI) in the urban area of Rome is
20 investigated in this study. In particular, the influence of aerosol optical depth (AOD) and single scattering
21 albedo (SSA), estimated at the wavelength of 340 nm, and of the Ångström exponent, calculated in the
22 range 340-500 nm, over a period of 11 years (2010-2020) in the months from March to September are
23 analyzed. The UVI is monitored by a Brewer spectrophotometer, whereas measurements of the direct
24 sun and diffuse sky irradiances are performed by a co-located PREDE-POM sun-sky radiometer of the
25 ESR/SKYNET network; the aerosol optical properties are obtained by the Skyrad MRiv2 retrieval. A
26 novel method, based on physical principles and easily adaptable to other contexts, is developed to
27 extrapolate the aerosol properties to the UV range during periods when only visible to near infrared
28 measurements are available. The retrievals from the sun-sky radiometer are consistent with the chemical
29 characterization of urban PM₁₀ (particulate matter 10 micrometers or less in diameter) samples, collected
30 during an intensive field campaign held in summer 2011 in the same site (URBan Sustainability Related
31 to Observed and Monitored Aerosol – URBS ROMA). The PM macro-components identified during the
32 campaign are grouped in order to evaluate the contribution of the main macro-sources (SOIL, SEA,
33 SECONDARY INORGANIC, ORGANICS and TRAFFIC) whose relative role is indeed expected to

34 strongly affect the aerosol absorption capability. The surface forcing efficiency, calculated as the change
35 in the UV index for a unit AOD variation, shows that AOD is the primary parameter affecting the surface
36 irradiance under clear sky conditions in Rome. SSA and the Ångström exponent are also identified as
37 secondary influencing factors, i.e., the surface forcing efficiency is found to be greater for smaller zenith
38 angles and for larger and more absorbing particles in the UV range (such as, e.g., mineral dust).

39

40 **1. Introduction**

41 The aerosol influence on the incoming and outgoing solar radiation is a widely studied topic because of
42 its relation with the Earth's radiative balance and climate. Indeed, the aerosol capability of attenuating
43 the UV radiation is responsible for its short and long-term variations and it has important implications
44 for tropospheric photochemistry, human health, and for the properties of organic materials, such as
45 plastics and wood routinely exposed solar radiation as well as aquatic systems (Dickerson et al., 1997;
46 He and Carmichael, 1999; Castro et al., 2001; Casasanta et al., 2011; Mok et al. 2018). Nevertheless,
47 various aspects are still uncertain because the columnar absorbing and scattering properties of
48 suspended particles are not very common in this wavelength region.

49 The aerosol optical depth (AOD) and single scattering albedo (SSA), that is the ratio of the aerosol
50 scattering to extinction coefficient, representing an index of the aerosol absorption capability, are
51 important radiative parameters to determine the aerosol effect on the UV irradiance at the surface.

52 Reuder and Schwander (1999) demonstrated that more than 80% of the aerosol effect on surface UV
53 radiation due to increasing turbidity of the atmosphere can be estimated through aerosol optical depth
54 and single scattering albedo. Strong UV absorption by aerosol, characterized by low SSA values at
55 wavelengths shorter than 400 nm, is commonly attributed to organic aerosols that absorb predominantly
56 in the UV region (Martins et al., 2009) and trigger a stronger wavelength dependence than for the pure
57 black carbon component (Kirchstetter et al., 2004). In effect, an enhancement of aerosol absorption at
58 UV wavelengths was observed in urban cities such as Rome, Italy (Ialongo et al., 2010) and Athens,
59 Greece (Kazadzis et al., 2016), especially in winter. Also, mineral components show a significant
60 absorption in the UV region, as highlighted by Meloni et al. (2006). Di Sarra et al. (2002), Panicker et
61 al. (2009), and Antón et al. (2011), among others, have shown that an increase of AOD induces a
62 reduction of the UV index (UVI), a relevant quantity expressing the levels of UV radiation potentially
63 harmful to human health. These studies suggested that a unit increase in aerosol optical depth at about
64 400 nm may produce a significant decrease of UVI which depends on the solar zenith angle and aerosol
65 properties, and may exceed 50%. More recently, Fountoulakis et al. (2021) showed that the positive
66 trends in UV irradiance detected in Rome in specific months during the last decades are possibly driven
67 by changes in clouds and/or aerosols.

68 This work is aimed at determining, for the first time in Rome, the effect of aerosol optical properties on
69 UV radiation, evaluating the role of SSA, AOD and Ångström exponent. Aerosol optical properties were
70 provided by a PREDE-POM sun-sky radiometer of the ESR/SKYNET (www.euroskyrad.net) network,
71 and the UVI values were obtained by UV irradiances measured by a Brewer spectrophotometer. The
72 dataset covers a period of 11 years, from 2010 to 2020, in the spring-summer months (March to
73 September). In the selected months, UVI at solar zenith angles (SZA) smaller than 40° are analyzed. For
74 $SZA > 40^\circ$, the uncertainty on the irradiances measured by the Brewer increases due to effects such as
75 stray light (Bais and Zerefos, 1996) and angular response error (Antòn et al., 2008). Therefore, an
76 enhancement of the estimated error of UVI, which is about 4-5%, (Schmalwieser et al., 2017) is also
77 expected. This could affect the identification of its variation caused by the aerosol effect.

78

79 **2. Data**

80 **2.1 Measurement site**

81 Rome is a large urban site, with about 3 million inhabitants, located 25 km east of the Tyrrhenian Sea,
82 in the middle of an undulating plain. The atmosphere is affected by urban emissions as well as by semi-
83 rural particulates and, especially during the spring and summer seasons, by sea breeze and long-range
84 desert dust advection from the Saharan region (e.g., Ciardini et al., 2012, Di Bernardino et al., 2021).
85 Long-term measurements of aerosol physical and optical properties, columnar ozone content and UV
86 irradiance (290 -325 nm) are carried out in Rome, on the roof of the Physics Department of Sapienza
87 University (41.9°N , 12.5°E ; altitude 75m), in the central sector of the city. This site since 2019 is part
88 of the BAQUNIN project (Iannarelli et al, 2021).

89

90 **2.2 Sun-sky radiometer measurements**

91 Aerosol properties are retrieved from observations taken in clear sky conditions by the sun-sky
92 radiometer PREDE/POM model 01 (hereafter called POM). It is a narrow band filter photometer able to
93 perform measurements of direct solar and diffuse sky irradiances within a narrow field of view of 1° at
94 selected wavelengths (315, 340, 400, 500, 675, 870, 940 and 1020 nm). The measurement schedule
95 includes direct irradiance observations every 1 minute and diffuse (almucantar, at 24 scattering angles
96 in the range $[0 -180^\circ]$) irradiances observations every 10 minutes. The 315 and 940 nm channels are
97 mainly used to retrieve ozone and water vapor columnar content, whereas the other ones provide
98 information on aerosols. The filter at 315 nm, which is characterized by a low transmittance, was
99 replaced with one centered at 340 nm in May 2016. This instrument is part of the European Skynet
100 Radiometer network (ESR, Campanelli et al., 2012; www.euroskyrad.net) that is a regional subnetwork
101 of SKYNET (Nakajima et al., 2021); it has been operating in Rome since 2010 up to present. Calibration

102 is performed monthly by the Improved Langley method (Campanelli et al., 2007), a well-tested “on-
103 site” procedure that allows to frequently check the instrument status.

104

105 **2.3 Brewer measurements**

106 Brewer MkIV spectrophotometer with the serial number 067 has been operating at the Solar Radiometry
107 Observatory of the Physics Department of Sapienza University of Rome since 1992. Total column ozone
108 have been recorded since 1992 whereas the spectral UV irradiance have been performed by the same
109 instrument since 1996. This instrument is part of the European Brewer Network (EUBREWNET) and
110 since 2019 has become part BAQUNIN project (Iannarelli et al, 2021).

111 The Brewer Mk-IV is a single monochromator spectrophotometer specifically designed to retrieve the
112 total column ozone by measuring solar direct irradiances at selected UV wavelengths in the ozone
113 absorption spectrum (Siani et al., 2018) and nitrogen dioxide in the visible part of the spectrum (Diémoz
114 et al., 2021). The accuracy of direct-sun measurements of total ozone taken with a well-maintained
115 Brewer spectrophotometer is 1% (Vanicek, 2006). The consistency among different instruments is ~1%
116 (Redondas et al., 2018). The Brewer also measures global spectral irradiances from 290 nm to 325 nm
117 with a step of 0.5 nm and a spectral resolution of about 0.6 nm. UV spectral scans are performed at Rome
118 every 30 min throughout the day. The performance of the Brewer instrument for UV measurements was
119 controlled every two years till 2014 through intercomparisons to the traveling reference QASUME UV
120 spectroradiometer operated by Physikalish Meteorologisches Observatorium Davos/ World Radiation
121 Centre (Gröbner et al., 2005). The mean ratio of Brewer integrated solar UV irradiances to QASUME is
122 within +3% (see <https://www.pmodwrc.ch/en/world-radiation-center-2/wcc-uv/>). Thereafter, the UV
123 calibration has been carried out by IOS using 1000 W lamps which are traceable to the National Institute
124 of Standard and Technology - NIST, Maryland, USA, and regularly compared to QASUME (Siani, et
125 al., 2013).

126

127 **2.4 Particulate Matter samples collected at the surface**

128 Results from an intensive field campaign (URBan Sustainability Related to Observed and Monitored
129 Aerosol – URBS ROMA, Campanelli et al., 2012) conducted in the period June – July 2011 in the same
130 location and aimed to determine the aerosol direct radiative effect at the surface, are also shown to
131 provide additional information on the aerosol properties at Rome during a summer season, and to
132 demonstrate the soundness of the column retrievals. PM₁₀ (particulate matter with diameter smaller than
133 10 micrometers) samples were collected by using a dual channel sampler (HYDRA Dual Sampler, FAI
134 Instruments, Fonte Nuova, Rome, IT) equipped with Teflon membrane filters and quartz fiber filters,

135 respectively on the two sampler's channels. The PM₁₀ mass concentration was measured on Teflon filters
136 by gravimetry using an automated microbalance.

137

138 **3. Methodology**

139 **3.1 Sun-sky radiometer retrieval method**

140 Skyrad_MRIv2. pack (Kudo et al., 2021), one of the official computer codes employed in the SKYNET
141 network, was used to retrieve the aerosol optical properties from the normalized radiance, which is the
142 ratio between the solar diffuse radiance and direct solar irradiance, at all available wavelengths. The
143 expected uncertainty in the retrieval products at near-ultraviolet, visible, and near-infrared wavelengths
144 is less than 0.04 for AOD, and less than 0.05 for SSA. The retrievals by Skyrad MRIv2 have been
145 previously validated based on in-situ measurements and radiative closure at the ground (Kudo et al.,
146 2021; Fasano et al., 2021a). A detailed comparison with the retrievals from a co-located AERONET
147 photometer operating at the BAQUNIN super-site is in preparation. The cloud screening was performed
148 based on the measurement of the fitness (f_{obs}) parameter introduced by Kudo et al. (2021). In order to
149 relate the UV Index to the aerosol optical properties, these latter have to be determined at a wavelength
150 as close as possible to the one corresponding to the maximum of the erythemally-weighted solar
151 spectrum (usually <320 nm, depending on the solar zenith angle). The shortest wavelength at which
152 reliable observations are available from the POM operating in Rome is 340 nm. Measurements at this
153 wavelength were started in 2016 ("dataset 2"), while the shortest wavelength was 400 nm prior to that
154 date ("dataset 1"). Hence, to increase the length of the aerosol dataset at the shortest measured
155 wavelength and cover a larger overlapping period with the UVI series from the Brewer, we developed a
156 new physically-based method (described below) to extrapolate the aerosol optical depth and aerosol
157 properties from longer wavelengths (400 nm and above) down to 340 nm for both dataset 1 and 2. Then,
158 to assess the accuracy of the method, we compare the outcome of this extrapolation with the retrieval
159 obtained using all available wavelengths, including 340 nm (period dataset 2). Based on the very good
160 results of such a comparison (Sect. 4.1), we always use the extrapolated data set throughout the period
161 analyzed here for consistency. Replacing the retrievals based on observations at 340 nm (in the few
162 periods when they are available) with the extrapolation is therefore not expected to affect the findings
163 of this study.

164 The extrapolation is performed using the Aerosol Optical Properties (AOP) program included in the
165 Skyrad MRIv2 package. AOP is able to calculate the aerosol optical properties at arbitrary wavelengths
166 from the results of the Skyrad inversion, using the same kernels as in the retrieval. For example, it is
167 normally used to interpolate the aerosol properties to the lidar wavelengths. The optical properties are
168 calculated by the procedure described by Kudo et al. (2021), i.e. from the retrieved volume size

169 distribution (VSD) and complex refractive index. The real (RRI) and imaginary (IRI) part of the
170 refractive index are interpolated in the log–log space. In the default AOP operation, the RRI and IRI at
171 the shortest/longest available wavelengths are used when properties at wavelengths beyond the measured
172 interval are requested. However, in the present study we further modified the code to allow extrapolation
173 outside the measurement interval. The limitations of this approach are discussed in the next sections.
174 Hence, the aerosol products considered in this study are: AOD_{340} , SSA_{340} , Absorption Aerosol Optical
175 Depth ($AAOD_{340} = AOD_{340} * (1 - SSA_{340})$) and Ångström exponent calculated from the AOD at 340 and
176 500 nm (Ae_{340_500}) to infer the AOD wavelength dependence in that spectral range.

177

178 **3.2 Brewer retrieval method**

179 Spectral UV irradiances, measured by the Brewer spectrophotometer in clear sky conditions (no clouds
180 obscure the sun), selected according to the Alexandrov et al. (2004) methodology, were used to retrieve
181 the UV index (UVI). The UVI was introduced in Canada in 1992 (Fioletov, 2010) to represent the
182 potentially harmful effects of UV radiation in a simple form. UVI is a unit-less quantity determined by
183 multiplying the erythemally weighted UV irradiances (in $W\ m^{-2}$) over the range 280-400 nm by $40\ m^2\ W^{-1}$
184 ¹ (COST-713, 2000). UVI values are grouped into exposure category expressing the risk for unprotected
185 skin to Sun exposure. Typically at mid-latitudes, UVI values at noon vary from 0 to 10, but highest UVI
186 values (a peak of 12.3 at Plateau Rosà, 3500 m a.s.l., in Valle d'Aosta Region, Italy) are experienced at
187 high altitudes (e.g., Casale et al., 2015) or at lower latitude sites.

188 The spectral irradiances were processed using the SHICrvm software version 3_075 (Slaper et al.,
189 1995) to check for any spectral wavelength shift and spectral anomalies (Slaper et al, 1995) in the UV
190 data. Furthermore, the software allowed to obtain the biologically effective UV irradiance by weighting
191 the solar irradiances with the erythemal action spectrum (C.I.E., 1999). Since the Brewer MKIV
192 spectrophotometer measures spectral irradiances up to 325 nm, the non-measured part of the UVA
193 spectrum needed for the calculation of UVI was extrapolated by the same software. Based on
194 considerations for similar corrections in the Brewer operating software (Fioletov et al., 2004), we
195 estimate an additional contribution (<2%) to the overall uncertainty in the UV index value for solar
196 zenith angles <70° due to this extrapolation.

197 Total ozone values (O_3) from direct-sun measurements were generated by using the Brewer Processing
198 Software, applying the rejection criteria on ozone values less than 100 DU and greater than 500 DU
199 (Siani et al., 2018). Yet, individual total ozone values were discarded when standard deviation is above
200 2.5 DU and ozone air mass is above 3.5 (the ozone air mass is defined as the ratio of the actual ozone
201 path length taken by the direct solar beam to the analogous vertical ozone path when the Sun is overhead
202 from the surface to the top of the atmosphere).

203

204 **3.3 PM samples chemical analysis**

205 Chemical characterization of the PM₁₀ samples collected during the URBS campaign was carried out
206 according to the method reported in Perrino et al. (2009). Briefly, elements were determined on Teflon
207 filters by X-ray fluorescence (XRF); then the filters were water-extracted and analyzed for their ionic
208 content by ion chromatography (IC); elemental and organic carbon (EC and OC) were detected on quartz
209 filters by thermo-optical analysis (NIOSH-QUARTZ temperature protocol). This overall analytical
210 procedure allows the determination of each individual component typically accounting for more than
211 1% of the PM₁₀ mass (macro-components: Si, Al, Fe, Na, K, Mg, Ca, chloride, nitrate, sulfate,
212 ammonium, elemental carbon, organic carbon) and to obtain the mass closure.

213 PM₁₀ macro-components can be grouped into five clusters to estimate the contribution of the main
214 macro-sources: SOIL, SEA, SECONDARY INORGANICS, ORGANICS, and TRAFFIC. Details about
215 the algorithms are reported in Perrino et al. (2014). The contribution of SOIL was calculated by adding
216 the concentration of those elements (considered as metal oxides) generally associated with mineral dust:
217 Al, Si, Fe, the insoluble fractions of K, Mg, and Ca (calculated as the difference between XRF and IC
218 determinations), calcium and magnesium carbonate (calculated as the sum of soluble calcium multiplied
219 by 1.5 and soluble magnesium multiplied by 2.5); SEA was estimated from the sum of Na⁺ and Cl⁻,
220 multiplied by 1.176 in order to take into account minor sea-water components; SECONDARY
221 INORGANICS were calculated as the sum of nitrate, ammonium and non-sea-salt sulphate; the
222 contribution of road TRAFFIC was estimated by adding elemental carbon to an equivalent amount
223 multiplied by 1.1 in order to consider the contribution of primary organic matter that can be adsorbed on
224 particles surface; the remaining organic carbon, multiplied by 1.6 to take into account non-C atoms,
225 constituted the ORGANICS and included both secondary organic species and primary components.

226

227 **3.4 Optical properties of surface aerosol from a radiative transfer model**

228 Rstar is a radiative transfer model (Nakajima and Tanaka 1986) able to simulate the radiation fields in
229 the atmosphere-land-ocean system in the wavelength range 0.17 – 1000 μm. Eight fundamental materials
230 (water, dust-like, sea salt, volcanic ash, yellow sand, ice, water-soluble, soot and 75%H₂SO₄) are
231 considered to assemble a three component internal mixture for each of the ten particles model types
232 (Water, dust-like, volcanic-ash, rural, urban, yellow sand, ice, soot, 75%H₂SO₄, sea spray, tropo). The
233 imaginary parts of refractive index of each fundamental material in the Rstar model were used as
234 reference values and helped to understand the variety of absorption capability of the PM₁₀ macro-
235 components concentration in the atmosphere over Rome.

236

237 3.5 Assessment of the dependence of UVI on the aerosol optical properties

238 To discern the dependence of UVI on the aerosol characteristics, the influence produced by solar zenith
239 angle total column of ozone, and orbital parameters (varying Earth-Sun distance) must be minimized.
240 Therefore, firstly the UVI was corrected for the variation of the Earth-Sun distance and values were
241 reduced to the mean Sun-Earth distance (Madronich, 1993). Secondly, only data at two values of
242 $SZA_{30^\circ} (\pm 1^\circ)$ and $40^\circ (\pm 1^\circ)$ were selected. This criterion excludes data related to autumn and winter
243 seasons, when the solar zenith angle is always higher than 40° in Rome. Thirdly, the UVI dependence
244 on total O_3 has been removed. This correction has been implemented using the Radiation Amplification
245 Factor (RAF) and scaling the UVI to the total ozone daily average value for the day with the lowest
246 AOD_{340} recorded in the entire dataset (283 DU on July 8, 2014). In fact, the effect of ozone on the
247 erythemal UV irradiance may be described as suggested by Madronich (1993) and Booth and Madronich
248 (1994):

$$249 \quad \frac{E^*}{E} = \left(\frac{O_3}{O_3^*} \right)^{RAF}, \quad (1)$$

250 where E and E^* are two UV irradiances observations, and O_3 and O_3^* their corresponding total ozone
251 amounts.

252 Similarly, it is possible to apply the above relationship to UVI:

$$253 \quad UVI^* = UVI \left(\frac{\langle O_3 \rangle}{O_3^*} \right)^{RAF}, \quad (2)$$

254 where $\langle O_3 \rangle$ is the daily ozone average value, O_3^* is the daily ozone average value during the day with
255 the minimum average AOD_{340} , and RAF is assumed to be equal to 1.25, according to di Sarra et al.
256 (2002). In that study the authors (Figure 8) retrieved values of RAF after correcting for the influence of
257 co-varying aerosol optical depth and they found the values between 1.0 and 1.2 at 30° and 40° solar
258 zenith angles when considering all aerosol conditions. As discussed in the paper, these values are
259 affected by different processes (the wavelength dependence of the aerosol sensitivity, the
260 interdependence between ozone and aerosol, possibly through increased ozone absorption following
261 enhanced scattering by aerosols, ozone and aerosol vertical distributions). The values of 1.25 was
262 derived from UVSPEC radiative transfer model calculations (Edme et al., 2016) where the aerosol
263 amount was kept fixed. This value is also in agreement with various other determinations of the ozone
264 RAF (e.g., De Luisi and Harris, 1983; McKenzie et al., 1991; Kerr and McElroy, 1993). However, a
265 sensitivity study of UVI^* on RAF variation from 1 to 1.25 has been performed over all the dataset
266 showing an average decrease of UVI^* by about 1.4%, that is within the declared uncertainty of 4-5%,
267 (Schmalwieser et al., 2017).

268 To point out the possible effect of aerosol optical characteristics measured at 340 nm on UVI*, AOD₃₄₀,
269 SSA₃₄₀, AAOD₃₄₀ and Ae₃₄₀₋₅₀₀ were analyzed as function of UVI* at the two fixed solar zenith angles,
270 associating estimations of aerosol parameters and UVI* made within ±5 minutes.

271

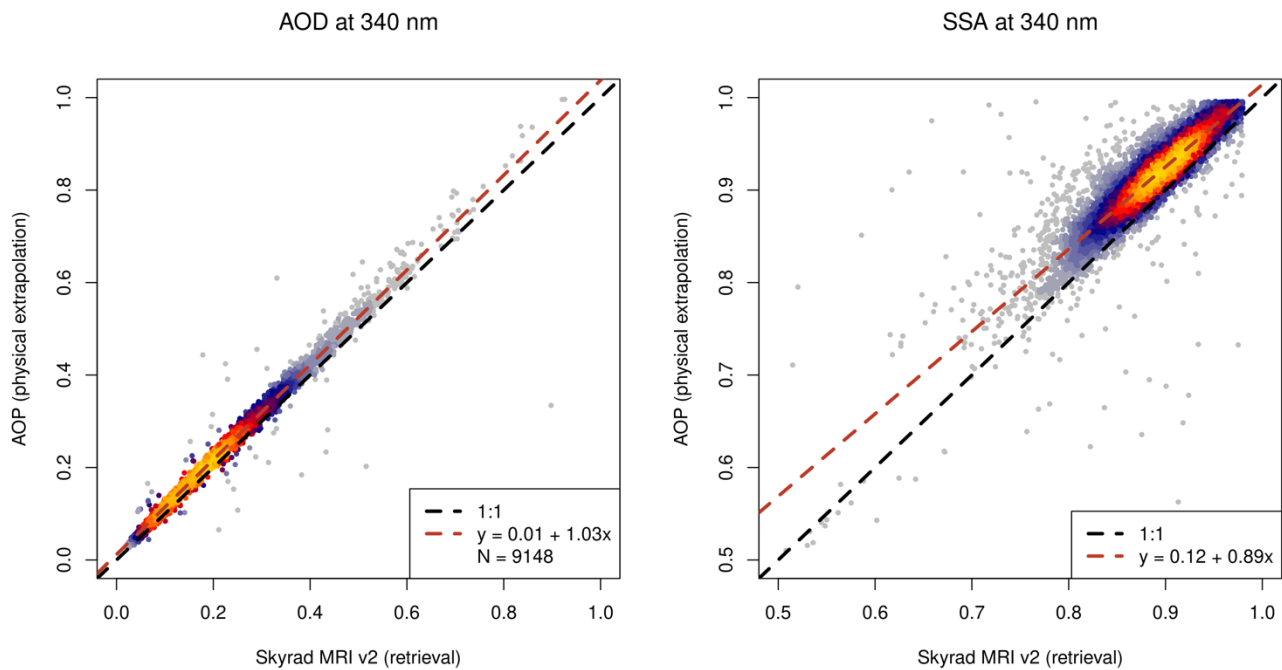
272 **4. Results**

273 **4.1 Validation of the method to extrapolate the aerosol properties to 340 nm**

274 Figure 1 shows a comparison of the AOD and SSA extrapolated at 340 nm by AOP (from wavelengths
275 of 400 nm and above) and retrieved by Skyrad MRIv2 using all wavelengths (including 340 nm), for the
276 period 2017-2020 (dataset 2). The discrepancies are well within the estimated uncertainties for both
277 AOD₃₄₀ and SSA₃₄₀. Regarding the latter, it is possible that the extrapolation leads to a slight
278 overestimation due to neglecting organic aerosols in the atmosphere, that are instead present in Rome as
279 found during the URBS campaign (Sect. 4.3). Indeed, the organic compounds are characterized by a
280 larger absorption in the UV than in the visible range (Massabò et al., 2015).

281 To remove this residual discrepancy, thus taking the likely effect of organics into account, we further
282 corrected the extrapolated data (AOD and SSA) by applying a simple offset/slope correction derived
283 from the linear regression of AOP (y) vs Skyrad (x, with $y = 0.12 + 0.89x$). By definition, this brings the
284 AOP results to overlap to the Skyrad MRI retrievals on average, for the selected period. This correction
285 assumes that the average relationship found for the 2017-2020 period also stands for the previous years.
286 To verify this hypothesis, we again compare this optimized extrapolation to the real Skyrad retrievals in
287 a different period (May-December 2016), when measurements at 340 nm are also available. Negligible
288 differences are obtained, on average, even for this period ($dAOD < 0.006$ and $dSSA < 0.01$, not shown),
289 hence demonstrating the robustness of the procedure.

290



291

292 Fig. 1: Comparison of the AOD (left panel) and SSA (right panel) at the wavelength of 340 nm retrieved
 293 from Skyrad MRIv2 using measurements at all wavelengths as input and the physically-based
 294 extrapolation from the visible-IR (>400 nm) to 340 nm based on the AOP program. Colors represent the
 295 density of the points based on a two-dimensional Kernel Density Estimation (Venables and Ripley,
 296 2013). Data refer to the period 2017-2020, i.e. a period when measurements in the 340-nm channel were
 297 available.

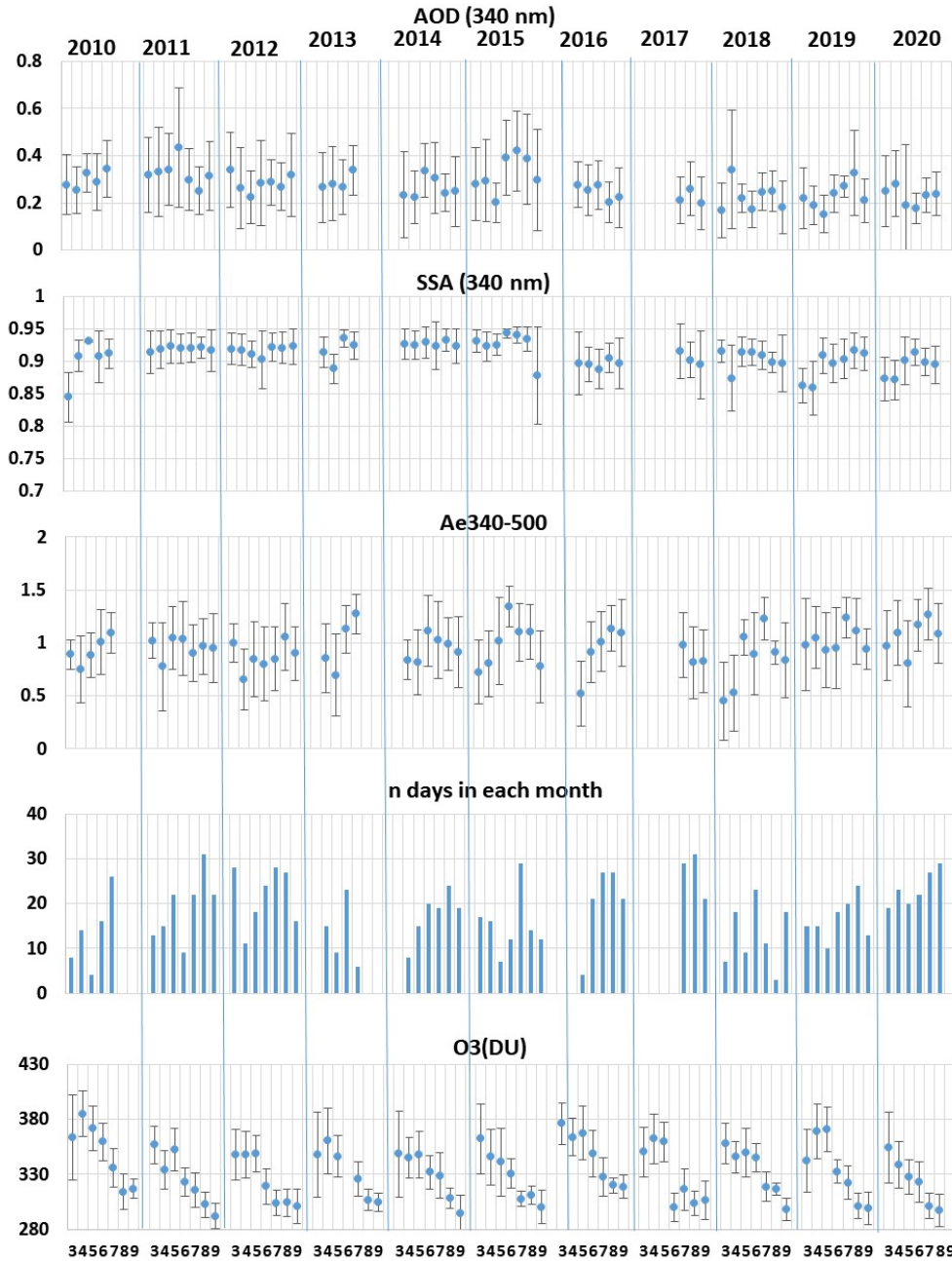
298

299 4.2 UVI dependence on aerosol optical parameters

300 The analyzed dataset covers the period March – September from 2010 to 2020 (for the last year the series
 301 ends in August). Figure 2 shows monthly averages of AOD₃₄₀, SSA₃₄₀, and Ae₃₄₀₋₅₀₀ for the period under
 302 study. Monthly means of SSA₃₄₀ vary between a minimum value of 0.84±0.04 (observed in March 2010)
 303 and a maximum of 0.94±0.01 (observed in June 2015). AOD₃₄₀ monthly mean values range between a
 304 minimum of 0.15±0.08 (in May 2019) and a maximum of 0.43±0.25 (in June 2015). Ae₃₄₀₋₅₀₀ varies
 305 between 0.45±0.37 (in March 2018) and 1.34±0.19 (in June 2015). The total ozone content values are
 306 also plotted in Figure. 2. The seasonal ozone behavior is typical of mid-latitude sites, with highest values
 307 measured in spring and particularly in April 2010 (385 DU) and the minimum in September 2011 (292
 308 DU).

309 During June-July 2011 the PM₁₀ chemical analyses show an average contribution over the entire sampled
 310 mass of about 29% of SOIL, 6% of SEA, 23% of SECONDARY INORGANIC, 28% of ORGANICS
 311 and 9% of TRAFFIC components. The absorption capability of these components is very different: in
 312 the Rstar radiative transfer model at 336 nm the imaginary part of refractive index varies from low values

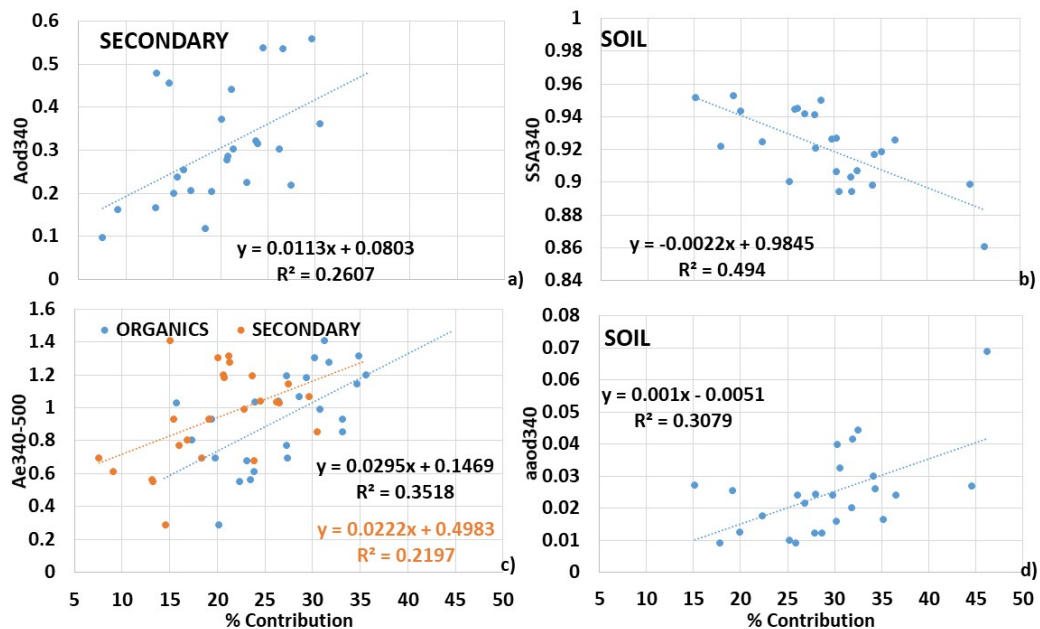
313 (weak absorption) of $4.02 \cdot 10^{-7}$ for marine aerosol (sea salt) and $1.00 \cdot 10^{-8}$ for 75% H_2SO_4
 314 (characterizing SECONDARY INORGANICS fraction) to a maximum of $4.70 \cdot 10^{-1}$ for soot
 315 (characterizing the TRAFFIC contribution). It is therefore expected that the modulation of the
 316 concentration of these co-existent materials, can strongly affect the absorption capability of the
 317 atmosphere over Rome.



318
 319 Figure 2: Monthly averages of AOD_{340} , SSA_{340} , $Ae_{340-500}$ and total O_3 for each year from 2010 to 2020.
 320 The number of points refers to the available daily mean data used in the calculation of the aerosol
 321 parameters monthly means. Error bars are the standard deviation that for SSA can be lower compared to
 322 the reported 0.05 error on SSA retrieval.

323

324 A regression analysis of the daily means of SSA_{340} , AOD_{340} , $AAOD_{340}$ and $Ae_{340-500}$ and the percentage
 325 contribution of each chemical component, has been performed in order to check whether the optical
 326 properties retrieved over the whole column and the chemical composition of samples collected at the
 327 surface are linked. Scatter plots of SSA_{340} and $AAOD_{340}$ versus the fraction of SOIL component
 328 contained in PM10 (Figure 3b, d) exhibit a reasonable correlation ($R^2= 0.49$ and 0.31 respectively)
 329 showing an increase of the absorption capability in the atmosphere (lower SSA_{340} and higher $AAOD_{340}$
 330 values) with the enhancement of the SOIL contribution. A weaker correlation is found between $Ae_{340-500}$
 331 and the percent amount of ORGANICS and SECONDARY INORGANICS contributions ($R^2= 0.35$
 332 and 0.22 respectively) highlighting that an enrichment of these particulates sampled at ground level may
 333 be associated with higher values of $Ae_{340-500}$ due to the presence of smaller particles in the atmosphere
 334 (Figure 4c). SECONDARY INORGANICS seems also to be weakly correlated with AOD_{340} in Rome.
 335



336
 337 Figure 3. Behaviour of AOD_{340} , SSA_{340} , $Ae_{340-500}$ and $AAOD_{340}$ versus the percentage contribution of
 338 some components as retrieved during the URBS campaign.

339
 340 In order to point out the possible effect of aerosol optical characteristics measured at 340 nm on UVI^* ,
 341 the AOD_{340} , SSA_{340} , $AAOD_{340}$, and $Ae_{340-500}$, were analyzed as function of UVI^* , at the two selected
 342 values of solar zenith angle. Figure 4 shows the frequency distributions of the number of measurements
 343 per month for each of the two angles. $SZA=30^\circ$ is more representative of the warmest months, whereas
 344 40° covers a wider period.

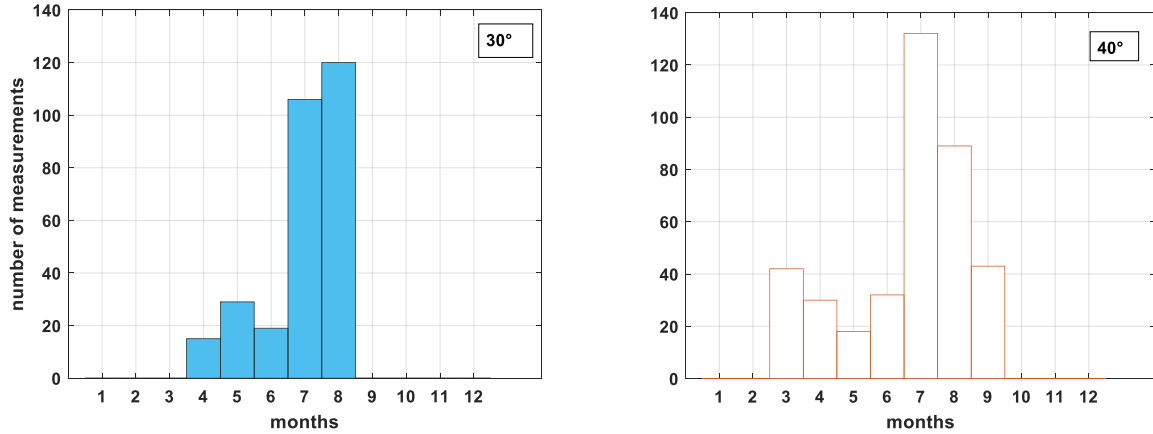


Figure 4: Number of measurements available for each zenith angle.

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348 The dependency of UVI^* on AOD_{340} , SSA_{340} , $AAOD_{340}$ and $Ae_{340-500}$ for 30° and 40° solar zenith angles
 349 is shown in Figure 5, colored for different values of SSA_{340} or AOD_{340} . Slopes, intercepts and correlation
 350 coefficients R^2 , are shown in Table I. The slope retrieved for UVI^* versus AOD corresponds to the UVI
 351 radiative forcing efficiency, i.e., the change in UVI produced by a unit change in AOD .

352 It is worthwhile mentioning that the UVI “effective” wavelengths are around 305-310nm depending on
 353 solar elevation, whereas aerosol properties are here retrieved at 340nm. Since AOD_{305} is theoretically
 354 higher than AOD_{340} for the same instant and the relationship between $AAOD_{305}$ and $AAOD_{340}$ depends
 355 on SSA spectral behavior in the UVB, the obtained results in Figure 5 and table I could be partly affected
 356 by the spectral behavior of both AOD and SSA in the 305-340 nm range. A slight decreasing trend of
 357 UVI^* when increasing AOD_{340} is evident at the smaller solar zenith angle, in agreement with what is
 358 found by previous studies (di Sarra et al., 2008; Antón et al., 2011). For the other parameters no
 359 dependence is visible from this first analysis. To investigate in more detail the possible effects on UVI^*
 360 caused by particles dimensions and atmospheric absorption capabilities, the entire dataset was divided
 361 in two groups of $Ae_{340-500}$ values, respectively below and above 1, and in two groups of SSA_{340} values,
 362 smaller and larger than 0.9, respectively. The values separating the different groups were determined
 363 according to the frequency distributions of the two variables for the entire investigation period, shown
 364 in Figure 6; they are the median values of the distributions. To understand if the extreme values of the
 365 distributions have a different impact on UVI^* , also the first and fourth quartiles were considered as
 366 thresholds values for the SSA_{340} and $Ae_{340-500}$ datasets: i) smaller than 0.8 and greater than 1.2, for $Ae_{340-500}$
 367 dataset; ii) smaller than 0.87 and larger than 0.93 for SSA_{340} . Scatter plots of UVI^* versus the two
 368 variables, for each group, were performed and points with a distance greater than 2σ from the regression
 369 line (nout), with σ the standard deviation of the residuals, were rejected for the final linear fit. The

370 dependence of UVI* on AOD for the classes of Ae₃₄₀₋₅₀₀ separated by median value, is shown in Figure
 371 7, colored for different values of SSA₃₄₀, and statistical results for all the classes are reported in Table I.
 372 At 30° the slope is greater for smaller values of Ae₃₄₀₋₅₀₀, both in the case of median and quartiles
 373 thresholds (-3.27 and -3.02 respectively), similarly to what found by Antón et al. (2011), and the
 374 correlation coefficients R² are the highest. Small values of Angstrom exponents are due to the presence
 375 of coarse particles, that in Rome are generally related to the presence of Saharan dust, more absorbing
 376 in the UV than VIS regions.

377 Smaller particles show a smaller forcing efficiency, despite the presence of organic aerosol likely
 378 absorbing in the UV range (Fig. 3). In fact in the case of Ae₃₄₀₋₅₀₀ greater than 1.0, which according to
 379 the analysis from the URBS campaign is likely related to concentrations from 15% to 35% of
 380 ORGANICS and SECONDARY INORGANICS fractions (Figure 3c), smaller values of slope (-1.28
 381 and -1.34) and R² are visible. These small particles are able to reduce the UVI* by about 1.0 when
 382 AOD₃₄₀ reaches values of 0.7. A similar dependency on the Ångström exponent was found by di Sarra
 383 et al. (2008) when considering the forcing efficiency over the whole shortwave spectral range.

384 At 40° SZA the slope is reduced with respect to 30°, as expected. Slightly larger (negative) slopes are
 385 found at 40° for Ae₃₄₀₋₅₀₀ values > 1.0, however these differences are not significant compared to the
 386 combined uncertainty of the slopes and might have been introduced by statistical sampling only.

387 Figure 8 shows the scatter plots of UVI* vs AOD₃₄₀ for SSA₃₄₀ < 0.9 (top) and SSA₃₄₀ ≥ 0.9 (bottom),
 388 with a colour scale for different values of the Ångström exponent at the two zenith angles. Less absorbing
 389 particles (SSA₃₄₀ > 0.9) shows similar slopes at 30° and 40°. The slopes increase in the case of a more
 390 absorbing atmosphere, in agreement with the findings by Antón et al. (2011) in Granada, Spain, where,
 391 as expected, stronger aerosol absorption leads to a large surface forcing efficiency. At 30°SZA slopes
 392 reach values of -2.68 and -2.52 in the case of median and quartiles thresholds, respectively. According
 393 to the results from the URBS campaign (Figure 4b, d) the absorption capability (SSA₃₄₀ and AAOD₃₄₀)
 394 in Rome is correlated to the increase of the SOIL concentration, therefore it could be inferred that the
 395 greater is this component in the atmosphere the larger is the radiative forcing efficiency. The slopes are
 396 again greater at 30° compared with the results at 40°.

397

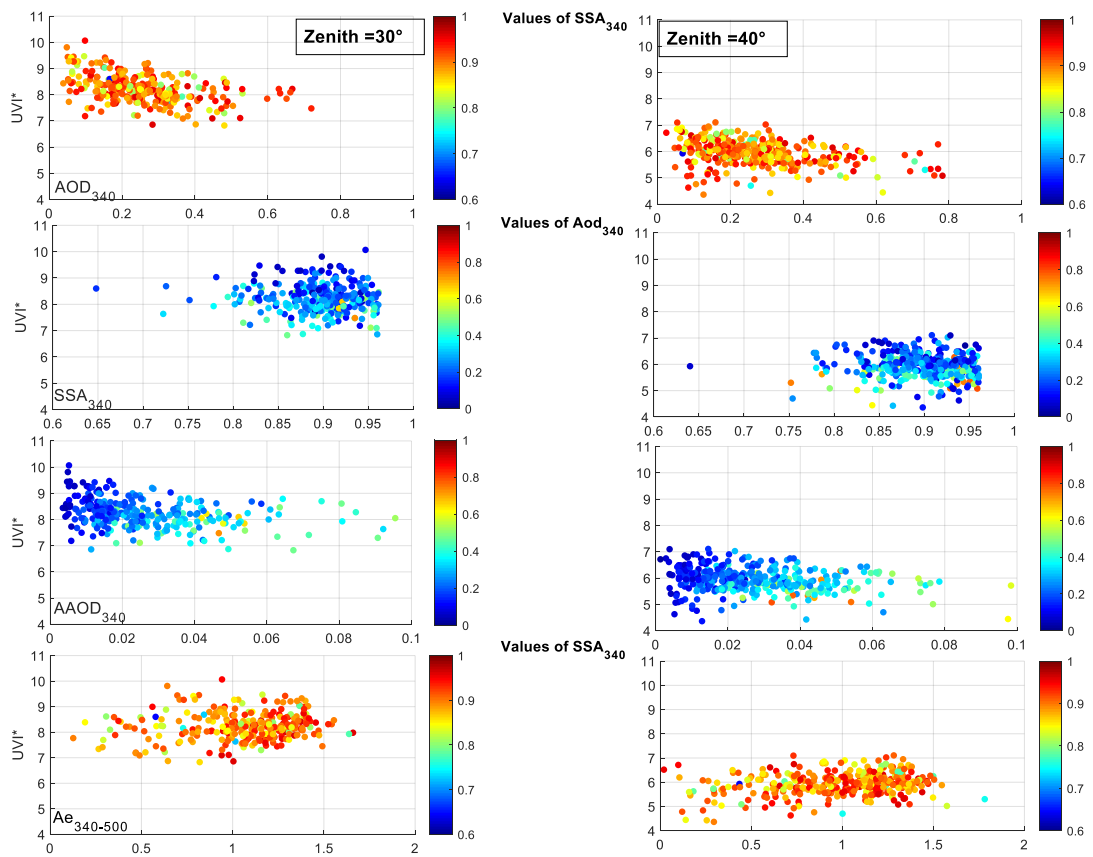
		SZA =30°			SZA =40°			
		UVI* vs:	Slope	Intercept	R ²	Slope	Intercept	R ²
	All data	AOD ₃₄₀	-1.91±0.23	8.72	0.21	-1.00±0.15	6.21	0.11
		AAOD ₃₄₀	-9.57±1.76	8.48	0.10	-5.51±1.12	6.09	0.06
		SSA ₃₄₀	-0.79±0.71	8.94	0.01	-0.66±0.53	6.52	0.004
	Ae ₃₄₀₋₅₀₀ < 1.0	AOD ₃₄₀	-3.27±0.32	8.87	0.58	-1.17±0.20	6.17	0.23

median	$Ae_{340-500} \geq 1.0$	AOD_{340}	-1.28 ± 0.18	8.55	0.27	-1.36 ± 0.12	6.37	0.41
quartiles	$Ae_{340-500} < 0.8$	AOD_{340}	-3.02 ± 0.35	8.68	0.66	-0.96 ± 0.25	6.07	0.15
	$Ae_{340-500} \geq 1.2$	AOD_{340}	-1.34 ± 0.22	8.63	0.33	-1.56 ± 0.15	6.49	0.55
median	$SSA_{340} < 0.9$	AOD_{340}	-2.68 ± 0.28	8.92	0.47	-1.65 ± 0.18	6.45	0.34
	$SSA_{340} \geq 0.9$	AOD_{340}	-1.08 ± 0.19	8.47	0.21	-1.05 ± 0.13	6.18	0.29
quartiles	$SSA_{340} < 0.87$	AOD_{340}	-2.52 ± 0.38	8.87	0.48	-1.83 ± 0.22	6.48	0.46
	$SSA_{340} \geq 0.93$	AOD_{340}	-0.97 ± 0.26	8.50	0.20	-0.78 ± 0.17	6.12	0.22

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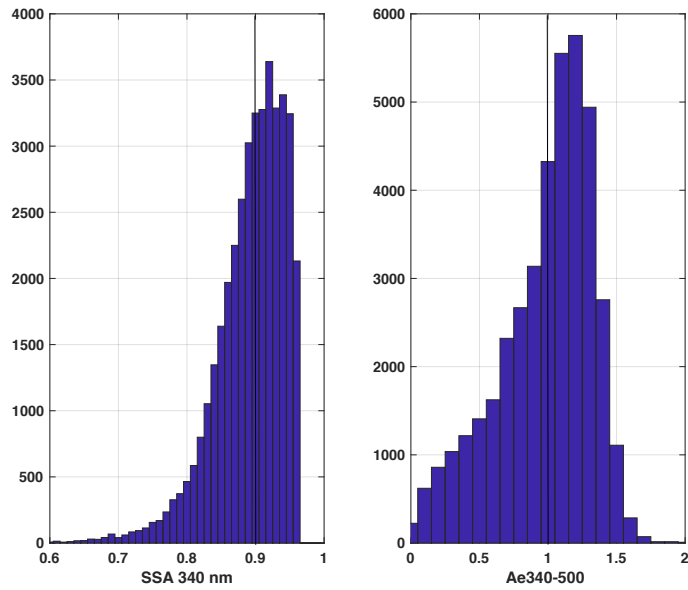
399 Table I: Slope, intercept and determination coefficient values for the linear fit of UVI* vs AOD_{340} ,
400 $AAOD_{340}$ and SSA_{34} for different cases and the two zenith angles

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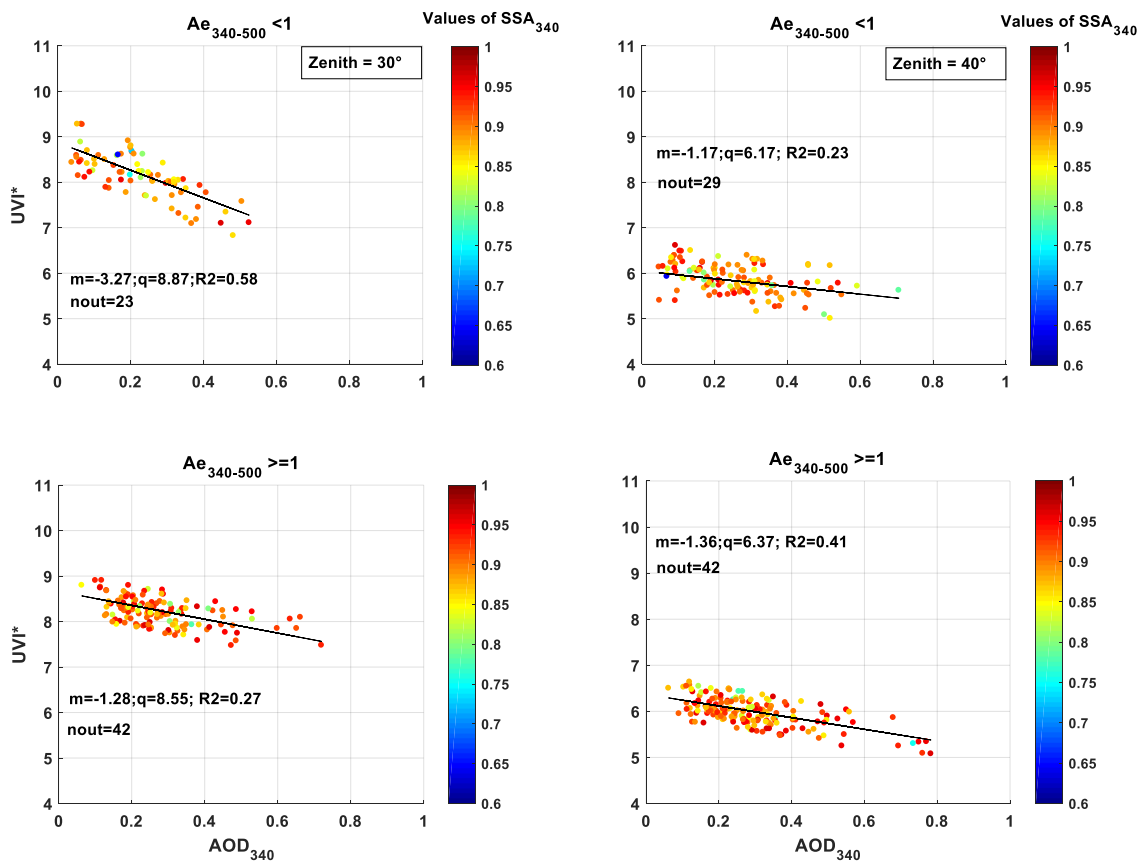
403 Figure 5 Scatter plot of UVI* vs AOD_{340} (top), SSA_{340} (second row), $AAOD_{340}$ (third row) and $Ae_{340-500}$
404 (bottom) for solar zenith angles of 30° (left) and of 40° (right). The colors represent the values of SSA_{340}
405 (first, and fourth rows) and AOD_{340} (second and third rows).



406

407 Figure 6. Frequency distributions of the values of SSA_{340} (left) and $Ae_{340-500}$ (right) for the entire
 408 investigation period. The median values used as threshold values to separate the different classes are
 409 highlighted with vertical black lines.

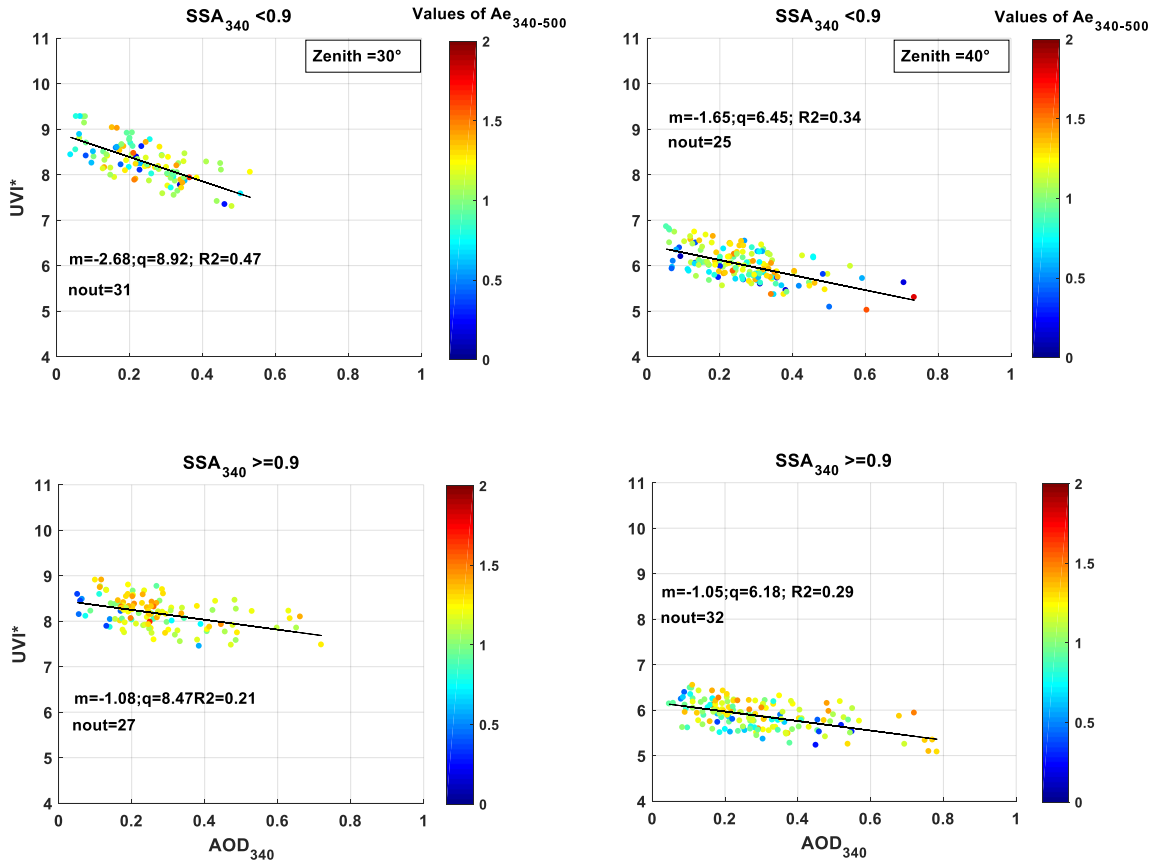
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411

412 Figure 7 scatter plot of UVI* vs AOD₃₄₀ for two groups of Ang₃₄₀₋₅₀₀ values and two solar zenith angles.
 413 The colors represent the values of SSA₃₄₀. “nout” is the number of rejected outliers. “m” is the slope and
 414 “q” is the intercept value.

415



416

417 Figure 8 scatter plot of UVI* vs AOD₃₄₀ for two groups of SSA₃₄₀ values and two solar zenith angles.
 418 The colour scale refers to the values of Ang. “nout” is the number of rejected outliers. “m” is the slope
 419 and “q” is the intercept value.

420

421 5. Conclusions

422 The aerosol optical properties in the urban area of Rome were retrieved for a period of 11 years, in the
 423 months from March to September 2010-2020, when also co-located surface measurements of the spectral
 424 UV irradiance were available. The impact of AOD and SSA at 340 nm, and Ångström exponent
 425 calculated at 340-500 nm on the UV Index has been analyzed. A new physically-based method was
 426 developed to extrapolate the aerosol optical properties to the UV range when only visible to infrared
 427 measurements were available. The method was validated based using data collected at all wavelengths
 428 and was applied to the period when measurements at 340 nm were not available. The dependence of
 429 UVI*, corrected for total ozone changes and scaled at the mean Sun-Earth distance, was studied with

430 respect to AOD_{340} , SSA_{340} , $AAOD_{340}$ and $Ae_{340-500}$. Data at two fixed values of the solar zenith angle
431 (30° and 40°) were selected in order to identify possible effects of the aerosol optical characteristics on
432 UVI^* . A slight decreasing trend of UVI^* when increasing AOD_{340} is evident at the smaller solar zenith
433 angle, with the slope corresponding to the UVI^* radiative forcing efficiency, i.e. the change in UVI^*
434 produced by a unit change in AOD. For the other parameters no dependence is visible.

435 To investigate in more detail the possible effects on UVI^* caused by particles dimensions and
436 atmospheric absorption capabilities, the entire dataset was divided in two groups of $Ae_{340-500}$, below and
437 above 1, and in two groups of SSA_{340} , smaller and larger than 0.9, where the thresholds are the median
438 values of the two data distributions.

439 The forcing efficiency was found greater for lower zenith angle and smaller values of $Ae_{340-500}$ both in
440 the case of median and quartiles thresholds. Small values of Angstrom exponents are related to the
441 presence of coarse particles, that in Rome are generally linked to the presence of Saharan dust in the
442 atmospheric column, more absorbing in the UV than VIS regions.

443 More absorbing particles ($SSA_{340} < 0.9$) showed a larger forcing efficiency at smaller zenith angles.
444 According to the results from the URBS campaign in Rome the absorption capability is correlated to the
445 increase of the SOIL fraction, therefore it could be inferred that the greater is this component in the
446 atmosphere the larger is the radiative forcing efficiency.

447

448 **Data availability**

449 All raw data can be provided by the corresponding authors upon request.

450

451 **Author contributions**

452 MC, HD and AdS wrote the manuscript draft; AdS MC, HD, AI, AS, GF, LT analyzed the data; MC,
453 AS, AI, GC, LT and MC, performed the measurements; RK developed the model and supported the
454 authors to its use; GC, LT, PS and SD reviewed and edited the manuscript.

455

456 **Competing interests**

457 The authors declare that they have no conflict of interest.

458

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