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Authors	Oliva, Fabrizio; D'AVERSA, EMILIANO; Liberti, G. L.; Sindoni, G.; ALTIERI, FRANCESCA; et al.
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A database of aerosols and gases coefficients for VIS-NIR radiative transfer in the Solar System planetary atmospheres.

F. Oliva ^{a,*}, E. D'Aversa ^a, G. L. Liberti ^b, G. Sindoni ^a, F. Altieri ^a, E. Castelli ^b

^aInstitute for Space Astrophysics and Planetology, INAF, Via del Fosso del Cavaliere 100, 00133 Rome, Italy

^bInstitute of Atmospheric Sciences and Climate, ISAC-CNR, Via Piero Gobetti 101, 40129, Bologna, Italy

Abstract

Radiative transfer (RT) modelling of planetary atmospheres allows to obtain extremely valuable information about these environments. The computation of optical properties of the aerosols and gases, that is heavily time consuming, is always required as input for both forward models and inversion algorithms. We provide look up tables for these coefficients, ready to be ingested in RT solvers, in the visible and near infrared spectral ranges and covering the main species of the Solar System planets' atmospheres. Gases absorption coefficients have been computed with the line-by-line approach from the HITRAN 2012 spectral database and an improved method to interpolate them is suggested. Aerosols single scattering properties have been computed using the Mie and Rayleigh models for a set of selected species relevant for planetary atmospheres. Optimization in terms of tabulated wavelengths and maximum number of Legendre polynomial expansion terms has been performed for each species.

Keywords: Radiative transfer
Spectroscopy
Aerosols optical constants
Gases absorption coefficients
Atmospheres, composition

1. Introduction

The modelling of radiative transfer (RT) is the main tool for interpreting remote sensing observations of planetary atmospheres (Haus and Arnold, 2010; Cess and Ramanathan, 1972; Määttänen et al., 2009; Geminale et al., 2015; Oliva et al., 2018; Karkoschka and Tomasko, 2005; Pérez-Hoyos et al., 2005; Oliva et al., 2016; Zhang et al., 2013; Sindoni et al., 2017; Chance and

* Corresponding Author. Tel: +39 06 45488201

E-mail address: (fabrizio.oliva@iaps.inaf.it)

37 Martin, 2017). Theories and models implementations in this field are developing since decades,
38 growing in accuracy to keep up with the increasing resolution of measurements in laboratory and
39 from space. Remote sensing in the visible (VIS) and near-infrared (NIR) spectral range is widely
40 used on almost all planetary exploration missions of current generation to probe composition (Titov
41 et al., 2007; Poulet et al., 2003; Fletcher et al., 2009), vertical structure (Sromovsky et al., 2013;
42 Pérez-Hoyos et al., 2012), thermodynamics (Fletcher et al., 2015; Colaprete and Toon, 2000), and
43 interactions of a planetary atmosphere (Brackett et al., 1995). Non-gaseous constituents of
44 atmospheres are often composed by particulates with typical sizes ranging from nanometre to
45 micrometres, i.e. of the same orders of magnitude of the wavelength of VIS and NIR light
46 (Karkoschka and Tomasko, 2005; Oliva et al., 2018; Sindoni et al., 2017). This, along with the
47 presence of important absorption complexes of the most diffuse molecules, makes this spectral
48 range preferred for studying the spatial and temporal distribution of particulates and their
49 microphysical properties (Oliva et al., 2018; Sindoni et al., 2017; Oliva et al., 2016; Sromovsky et
50 al., 2013, Roman et al., 2013).

51 The retrieval of physical quantities from observations is highly dependent on the accuracy of
52 our modeling of light absorption and scattering processes taking place within the different
53 atmospheric layers. Wavelength-dependent absorption and scattering coefficients (hereafter Optical
54 Properties, OP) are commonly used to parameterize the propagation of the radiation through the
55 atmosphere, as they can be evaluated separately for each gaseous and solid/liquid component in
56 their own thermodynamic condition. Therefore, the OPs represent the bulk of the atmospheric
57 models used for both forward and inverse RT computations. However, their evaluation with
58 sufficient accuracy is often heavily time consuming, also because they have to be evaluated for all
59 possible conditions of temperature, pressure, and composition expected in the probed atmospheric
60 layers.

61 As far as the gases absorption is concerned, the most rigorous approach for computing
62 absorption coefficients for any temperature and pressure is the line-by-line (LBL) one. It requires
63 the calculation of the absorption distribution in frequency for each molecular roto-vibrational
64 transition in a given spectral range, and allows taking into account several line broadening
65 processes. Since the line broadening is the effect of several pressure- and temperature-dependent
66 processes, the computational load of LBL is essentially related to the repeating evaluation of the
67 line shape at least for each molecular line and different pressure, temperature, and mixing ratio
68 values.

69 Several approximated methods have been proposed to speed up the computations. For
70 example, band models have been developed in the past for parameterizing the variations of
71 individual molecular absorption bands with pressure and temperature (e.g. Goody, 1952; Pierrot et
72 al., 1999; Karkoschka and Tomasko, 2010), but they suffer strong limitations and low accuracy for
73 atmospheric spectroscopic studies. The correlated-k technique (Goody and Yung, 1989), based on
74 the statistical distribution of absorption coefficients within suitable spectral bins, demonstrated
75 better performances in reproducing gaseous absorption for planetary atmospheres (Tsang et al.,
76 1999; Irwin et al., 2008; West et al., 2010). However, it is strongly constrained in terms of spectral
77 resolution, since computation cannot be done for intervals smaller than the initial bins, and the
78 computational cost greatly increases when the bins become narrower and narrower.

79 In order to keep application flexibility in terms of spectral resolution for all extra-terrestrial
80 planetary atmospheres, in this work we adopted the most rigorous LBL approach, linked with an
81 interpolation scheme applied to a look up table (LUT) (Section 2.2). Attempts to interpolate LUTs
82 of absorption coefficients have been made by other authors (e.g. Hill et al., 2013) with results that
83 vary with the application field. In this framework, an improved interpolation method is tested.

84 As a starting point for defining the working ranges of OP databases we take into account the
 85 characteristics of existing VIS to NIR spectral datasets about different Solar System atmospheres
 86 taken by space-based spectrometers (see Table 1). These datasets include extremely valuable
 87 information about the atmospheres of Mars, Venus, Jupiter, Saturn, and Titan, that will be still
 88 under investigation for years. We take also into account features of future spectrometers like MAJIS
 89 (Piccioni et al., 2014), that will fly on board JUICE mission towards Jupiter’s system. Hence, this
 90 choice drives the definition of the overall framework for evaluating optical properties. In fact, we
 91 only focused on the main absorbing molecules in planetary atmospheres, i.e. CH₄, CO₂, and H₂O,
 92 within the regimes of temperatures, pressures, and abundances probed by remote sensing in the
 93 considered planets. The spectral range for OP evaluation has been chosen to lie from 400 to 5500
 94 nm, as a compromise between the ranges covered by selected spectrometers and those where
 95 molecular transitions and refraction indices are provided in literature.

96 **Table 1: Definition of application fields for the considered datasets of OP ((a) Brown et al., 2004; (b)**
 97 **Bibring et al., 2005; (c) Piccioni et al., 2007; (d) Adriani et al. 2017; (e) Piccioni et al., 2014.)**

Instrument	Spectral range [μm]	Spectral res [nm]	atmosphere	Main absorber	T range [K]	P max [bar]
Cassini VIMS (a)	0.35-5.1	7-17	Saturn	CH ₄	80-220	~ 2.5
			Titan	CH ₄	70-170	~ 1.4
MEx OMEGA (b)	0.35-5.1	13-20	Mars	CO ₂ H ₂ O	150-270	~ 0.006
VEx VIRTIS (c)	0.3-5.0	1.8-9.8	Venus	CO ₂	170-580	~ 90
Juno JIRAM (d)	2.0-5.0	9-12	Jupiter	CH ₄	110-270	~ 2.5
Juice MAJIS (e)	0.4-5.7	2.3-6.6				

98 Another heavily time consuming operation in RT modelling is the estimation of Single
 99 Scattering Optical Properties (SSOPs) especially for particles that are relatively large compared to
 100 the wavelength at which the simulation is done. This issue is even more important in inversion
 101 algorithms that make use of real-time SSOPs computations. Radiative effects of condensate matter
 102 in planetary atmospheres cannot be neglected in the UV-NIR range. Retrieval of clouds and
 103 aerosols properties is a primary scientific subject to understand atmospheric processes (e.g. Oliva et
 104 al. 2016). Furthermore, surface studies take great advantage of the realistic representation of the
 105 condensate SSOPs, since it allows to remove the particulates contribution from the measurements
 106 (Brown, et al., 2014; Geminale et al., 2015). For these reasons, RT modeling should include a
 107 computationally efficient but accurate representation of the SSOPs. A way to achieve this is by
 108 storing the values of SSOP in pre-computed LUTs (Buehler et al., 2011; Vincent and Dudhia,
 109 2017).

110 In this study we adopt the LUTs approach for both gas absorption coefficients (GAC) and
 111 particulates single scattering parameters (SSOP). In Section 2 we describe the techniques and
 112 approximations applied to the computation of GAC, whereas the Section 3 is devoted to SSOP
 113 description. Some application of LUTs to forward RT models and to spectral inversion are
 114 described in Section 4, and future perspective are illustrated in Section 5.

115 The full database, composed by several data files of very large dimensions (some gigabytes),
 116 is made available through ESA Planetary Science Archive (PSA) accessible at the website
 117 <https://archives.esac.esa.int/psa/>. Moreover, any specific request on the database can be made to the

118 authors.

119 2. Gaseous Absorption Coefficients

120 As abovementioned, we adopted the rigorous LBL approach to compute GACs for CH₄, CO₂,
121 and H₂O molecules, by taking advantage of the ARS RT libraries (Ignatiev et al., 2005). The
122 spectroscopic line parameters for these molecules have been taken from the HITRAN 2012
123 database (Rothman et al., 2013). This database lists a total amount of more than 800000 lines for
124 these molecules in the 400-5500 nm range.

125 2.1. Absorption coefficients calculations

126 Absorption coefficients for a given molecular species at wavenumber ν , pressure P ,
127 temperature T , and mixing ratio χ is evaluated by LBL method as (Rothman et al., 1998):

128

$$129 \quad k(\nu, P, T) = \sum_{j=0}^N S_j(T_0) f_j(\nu, \nu_{0,j}, P, T, \chi) \frac{Q(T_0)}{Q(T)} e^{-\frac{hc}{k_B} E_j \left(\frac{1}{T} - \frac{1}{T_0} \right)} \left(1 - e^{-\frac{hc\nu_{0,j}}{k_B T}} \right) / \left(1 - e^{-\frac{hc\nu_{0,j}}{k_B T_0}} \right) \quad (1)$$

130

131 where the sum is extended to N molecular transitions of the species in a wide enough range, S_j is the
132 line strength of j^{th} transition centered at wavenumber ν_0 and evaluated at a reference temperature T_0 ,
133 and f_j and E_j are the broadened line profile and the lower state energy of that transition respectively,
134 Q is the sum partition function, h , c , and k_B are the Planck constant, light speed, and Boltzmann
135 constant respectively.

136 It is evident from (1) that the spatial and temporal variations of gaseous absorption in a real
137 atmosphere are made complicated by to the highly non-linear dependence of these coefficients on
138 pressure, temperature and mixing ratio.

139 In the present work, we compute the absorption coefficients over a three-dimensional grid in
140 the space of parameters, defined by all the combinations of the values given in Table 3: 10 values of
141 pressures, logarithmically spaced between 5 bars and 10^{-6} bars, 10 values of temperatures, almost
142 linearly spaced between 70 K and 500 K, and 4 values of mixing ratio spanning several orders of
143 magnitude. Therefore, the total number of grid points amounts to 400 values for each wavelength.

144 We adopted the Voigt line profile to describe the broadened shape of individual molecular
145 lines, taking into account thermal and collisional broadening processes. The Lorentzian broadening
146 halfwidths (α) are calculated following Rothman et al. (1998):

147

$$148 \quad \alpha(P, T, \chi) = [\alpha_{air}(P_0, T_0)(1 - \chi) + \alpha_{self}(P_0, T_0)\chi] \frac{P}{P_0} \left(\frac{T_0}{T} \right)^m \quad (2)$$

149

150 where α_{air} and α_{self} are the air- and self-broadening halfwidths, m is the exponent of temperature
151 dependence, P_0 and T_0 indicate reference values of pressure and temperature, and χ is the mixing
152 ratio of the species. Since α_{air} and α_{self} are not always available for the atmospheres of the Solar
153 System planets, we assumed the terrestrial values given in the HITRAN 2012 database.
154 Furthermore, isotopic fractionation is also kept on terrestrial values. Also the temperature
155 dependence of the Q functions has been taken from HITRAN 2012.

156 Since line shapes are computed over a same regularly-spaced wavenumbers grid for all lines,
157 we searched for an optimal wavenumber step allowing a trade-off between the accuracy on line

158 profile and the amount of data volume and calculation time. As a consequence, for each molecule
 159 we define spectral subintervals where grid steps can be kept constant, switching its value from 10^{-4}
 160 to 10^{-2} cm^{-1} based on lines density and broadening parameters (see Table 2). The errors we make
 161 with respect to a finer case (e.g. 10^{-5} cm^{-1}) are on average of the same order of the uncertainties due
 162 to our interpolation method (see Section 2.2 and Figure 2). For this reason, we discarded the
 163 possibility to use the finer case, since the enhancement in precision that it provides is not worth the
 164 increase in computational time and storage.

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166

167

168 **Table 2: spectral subinterval used for line shape computations. δ represents the wavenumber step**
 169 **adopted for line broadening calculations. Bold numbers indicate total values.**

CH4			CO2			H2O		
Intervals (cm^{-1})	Lines number	δ (cm^{-1})	Intervals (cm^{-1})	Lines number	δ (cm^{-1})	Intervals (cm^{-1})	Lines number	δ (cm^{-1})
870-1250	29124	10^{-2}	782-1200	7869	10^{-2}	388-1030	92931	10^{-3}
1250-3060	176661	10^{-3}	1200-1850	66540	10^{-3}	1030-5999	106289	10^{-4}
3060-4890	96606	10^{-4}	1850-3470	170054	10^{-4}			
4890-6000	12853	5×10^{-4}	3470-3950	7383	10^{-3}			
			3950-5951	99576	10^{-4}			
870-6000	315244		782-5951	351442		388-5999	199220	

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176 The wavenumber step used for shaping individual lines yields absorption coefficients
 177 guarantees the applicability of the LUTs to the analysis of the datasets in Table 1 (as well as of
 178 other similar datasets), being much narrower than the maximum resolution of these instruments
 179 (e.g. 1.8 nm/band of the visual channel of VIRTIS-M).

180 The HITRAN database also provides an estimation of uncertainties associated to each line
 181 parameter, which propagate non-linearly into the LBL calculations. We computed the uncertainties
 182 associated to absorption coefficients by using analytical derivatives of the line profile in the
 183 Lorentzian approximation. Average errors due to uncertainties on line strengths only, obtained in
 184 the spectral range under study in different P and T conditions, vary between 5% and 30% (e.g. left
 185 panel of Figure 2 for CH_4).

186

187 2.2. Absorption coefficients interpolation

188 We tested the application of several interpolation methods to the LUTs to check their
 189 usefulness in providing absorption coefficients for intermediate values of P , T , and χ . These

190 methods include fits of the absorption coefficients or their logarithm, using different degree
 191 polynomials. These fits were performed on: P, T and χ ; their logarithm; their inverse. The most
 192 performing method, that is more accurate in reproducing the values of exact calculations, relies on a
 193 three-steps procedure: 1) 7th degree logarithmic polynomial fit with respect to P; 2) 7th degree
 194 polynomial fit of the fit coefficients obtained in step 1 with respect to the inverse of T; 3) 2nd degree
 195 polynomial fit of the fit coefficients from step 2 with respect to χ . We used a 7th degree polynomial
 196 for steps 1) and 2) because lower degrees are not as precise (right panel of Figure 2) and higher
 197 ones do not provide an improvement that is worth the increase in computational time and storage.

198 The scheme of this procedure is shown in Figure 1. Step 1 follows the green arrow direction by
 199 interpolating the logarithm of the absorption coefficients over the logarithm of pressures. Step 2
 200 follows the yellow arrow direction interpolating the fit coefficients from step 1 over the inverse of
 201 temperatures. Finally, step 3 follows the red arrow interpolating the fit coefficients from steps 2
 202 over the mixing ratios.

203 The uncertainty between the exact CH₄ absorption coefficients computed with $P=162$ mbar, $T=200$
 204 K, $\chi =0.001$ and the interpolated ones is shown in Figure 2. In the whole spectral range considered
 205 for methane (870 nm – 6000 nm, left panel), the interpolation method from this work produces
 206 average uncertainties of about 5% (black line). By applying our method to different sets of the P,T
 207 and χ values given in Table 3 we verified that the above average errors represent an upper limit,
 208 demonstrating the reliability of the procedure. In the same range, the average uncertainties from Hill
 209 et al. (2013) interpolation method, adopted in the τ -REX RT tool (Waldmann et al., 2015), are
 210 about 35% (green line). As reference, the errors computed from the line intensity uncertainties
 211 given in the HITRAN database oscillate between 5% and 30% (orange line). The right panel of
 212 Figure 2 provides a zoom in the small range 870.00 nm – 870.05 nm to better show the
 213 uncertainties deriving from different interpolation methods.

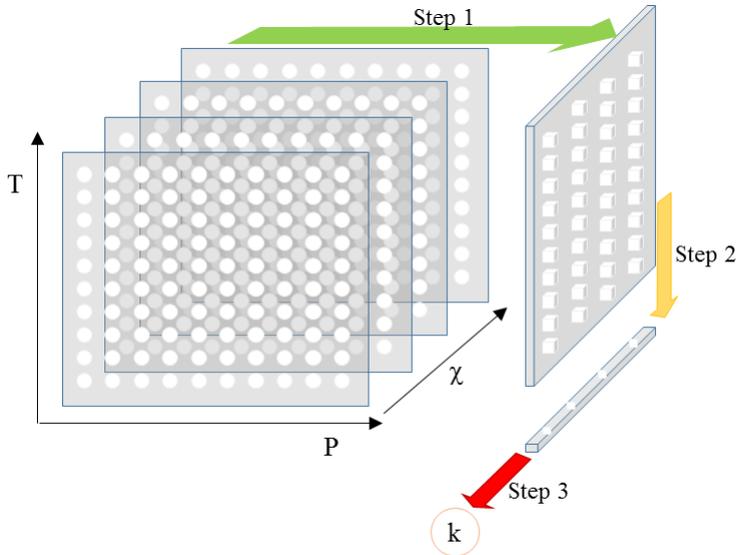
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T [K]	P [mbar]	χ
70	0.001	10 ⁻⁵
100	0.006	10 ⁻⁴
150	0.03	10 ⁻³
200	0.2	1
250	1	
300	5	
350	30	
400	160	
450	900	
500	5000	

217 **Table 3: values of T (panel A), P (panel B)**
 218 **and χ (panel C) adopted for the**
 219 **computations. All combinations of the**

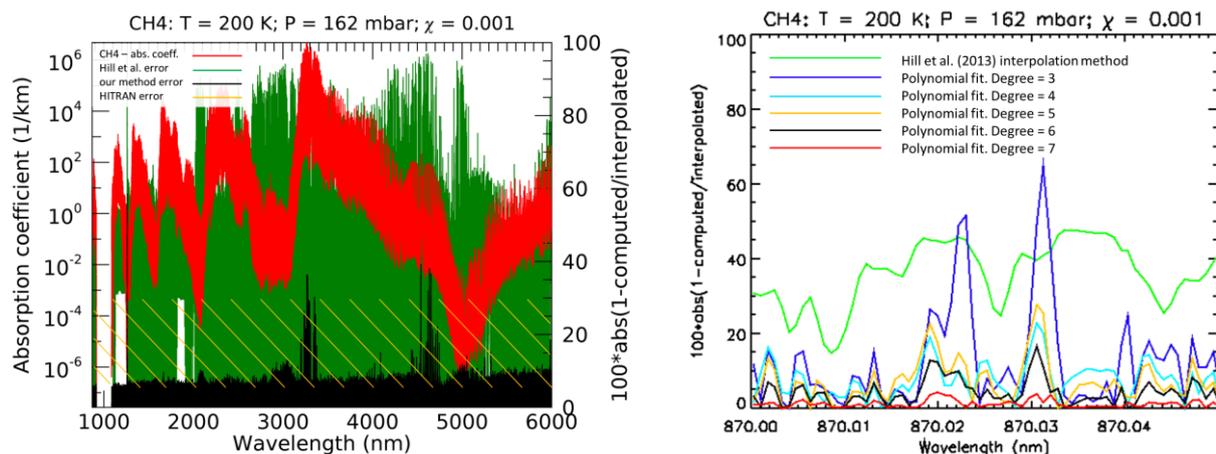
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values in panels A, B and C have been considered.



223
224
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226
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Figure 1: Representation in the space of the parameters of the 3-steps interpolation scheme described in Section 2.2 to obtain the absorption coefficients at the desired T and P for a given value of χ .



228

229 **Figure 2: At left:** relative differences (right axis) between CH₄ absorption coefficients computed with P
 230 = 162 mbar, T = 200 K, $\chi = 0.001$ between 870 nm and 6000 nm (red line, left ordinate axis) and those
 231 interpolated with our method (black line) and with that from Hill et al. (2013) (green line). The orange
 232 oblique lines indicate the range of uncertainty due to HITRAN line intensity error (Section 2.1). **At**
 233 **right:** zoom in the range 870.00 nm – 870.05 nm of the relative differences between the computed
 234 absorption coefficients (left panel, red line) and the ones interpolated with polynomial fits of different
 235 degree: 3rd degree (blue line), 4th degree (cyan line), 5th degree (orange lines), 6th degree (black line)
 236 and 7th degree (red line). The relative difference obtained with Hill et al. (2013) interpolation method is
 237 shown in green.

238 3. Aerosols optical properties

239 3.1. Refractive indices data

240 Single scattering optical properties (SSOPs) were computed for the most common ices and
 241 aerosols found in the Solar System atmospheres taking advantage of the optical constants available
 242 in literature. The considered species are given in Table 4. Mars dust (Wolff et al., 2009), is mainly
 243 composed by nanophase ferric oxide particles (Morris et al., 2006; Poulet et al., 2007) and is
 244 produced by soil weathering (Pollack et al., 1979; Korablev et al., 2005), strongly affecting the
 245 planet’s thermal structure and climate (Forget et al., 1999). Tholins (Khare et al., 1984; Cruikshank
 246 et al., 1993) are mixtures deriving from the photochemistry of molecular nitrogen and methane (N₂-
 247 CH₄) and are candidate to be the main component of Titan aerosols. H₂SO₄ clouds (Knollenberg &
 248 Hunten, 1980) dominate Venus cloud deck, likely of volcanic origins (Brackett et al., 1995). NH₃
 249 and NH₄SH ices (Howett et al., 2007) are expected as condensed clouds on Jupiter and Saturn. CO₂
 250 ice (Hansen et al., 2005) is known to condense in clouds in Martian atmosphere (Montmessin et al.,
 251 2007). HCN ice (Moore et al., 2010) is known to be a condensable species in Titan’s atmosphere.
 252 H₂O ice (Mastrapa et al., 2008, 2009) is the most common ice in the Solar System.

253

254 **Table 4: Summary of the solid/liquid species for which single scattering properties are calculated.**

Species	T (K)	Particle effective radius		Spectral range		N _{max}
		$r_{min}(\mu m)$	$r_{max}(\mu m)$	$\lambda_{min}(nm)$	$\lambda_{max}(nm)$	
Mars dust (Wolff et al., 2009)	-	0.1	10	263	4994	917
N ₂ CH ₄ Titan Tholin (Khare et al., 1984)	-	0.1	50	1842	5464	639
H ₂ SO ₄	-	0.1	10	250	5494	950

(Knollenberg & Hunten, 1980)						
NH ₃ ice (Howett et al., 2007)	80	0.1	20	840	5494	541
NH ₄ SH ice (Howett et al., 2007)	80; 160	1.0	100	833	5494	2493
H ₂ O ice (Mastrapa et al., 2008,2009)	80; 120	0.01	50	1111	5500	1076
CO ₂ ice (Hansen et al., 2005)	-	0.01	10	252	5500	94
HCN ice (Moore et al., 2010)	95	0.1	10	2000	4033	117
Triton Tholin (Cruikshank et al., 1993)	-	0.1	50	258	740	4643

255 3.2. Computation of scattering Look-Up tables

256 In this section we describe the strategy adopted to produce and optimize the SSOPs LUTs. Firstly,
257 the following assumptions were adopted:

- 258 - Particles are spherical: this assumption, relevant only for size parameter ($x=2\pi r/\lambda$) for which
259 the Rayleigh approximation is no more valid, allows the adoption of the Mie theory for
260 which well documented and relatively efficient software for the computation of SSOPs are
261 available;
- 262 - Particles are homogeneous and only external mixing is allowed;
- 263 - Simulations are performed with a scalar RT Model. This assumption, that allows to
264 represent the scattering matrix only with the description of the phase function, derives from
265 the characteristics of the instruments for which the radiative transfer model is developed.
266 These have very low sensitivity to the polarization of the measured radiances.

267 With the above assumptions, at a given wavelength, a single particle will be fully described
268 by the value of its radius and of the two components of the complex refractive index. In terms of
269 stored SSOPs the particle is characterized by the value of its extinction cross section, the single
270 scattering albedo and a number of un-normalized coefficients for the Legendre polynomials. This
271 approach has been preferred because most of the RT models require the Legendre coefficients as
272 input for the phase function computation.

273 SSOPs were computed assuming spherical particles and taking advantage of the MIEV0 program
274 (Wiscombe, 1980). Hence, the maximum extent of particle radius values was defined based on the
275 limits of MIEV0 within the spectral range of interest, that is constrained in terms of size parameter.
276 No attempt was done to estimate SSOP for size parameter values larger than the upper limit for
277 which the validity of MIEV0 software was tested ($x = 20000$). On the opposite edge, Mie model has
278 not been applied below the limit $x = 0.1$. In this case we tested several small-particles
279 approximations (e.g. Penndorf 1962), finally adopting the Rayleigh one in the SSOP LUTs. It is
280 worth noting that small-particles approximations represent a computationally efficient alternative to
281 the Mie theory within their validity range, and in this case retrieval algorithms can better exploit a
282 real-time evaluation of SSOP instead of resorting to LUTs. However, no attempt was done to
283 optimize the computation of SSOP in the Rayleigh-Mie transition region ($0.1 \leq x \leq 2$).

284 From the above considerations, for a given species, the dimension of the stored LUTs will depend
285 from the number of wavelengths, particles radii and Legendre coefficients.

286 For practical reasons, the approach of a fixed number of radii and maximum number of Legendre
287 coefficients was adopted. SSOPs were computed for a set of 1366 values of radii ranging from
288 0.001 to 791.89 μm and each differing of 1% with respect to the previous value.

289 Since we have a fixed number of radii, the optimization of the size of the LUTs was based on the
290 reduction of the number of wavelengths at which SSOPs are tabulated and on the definition of the
291 minimum number of Legendre coefficients.

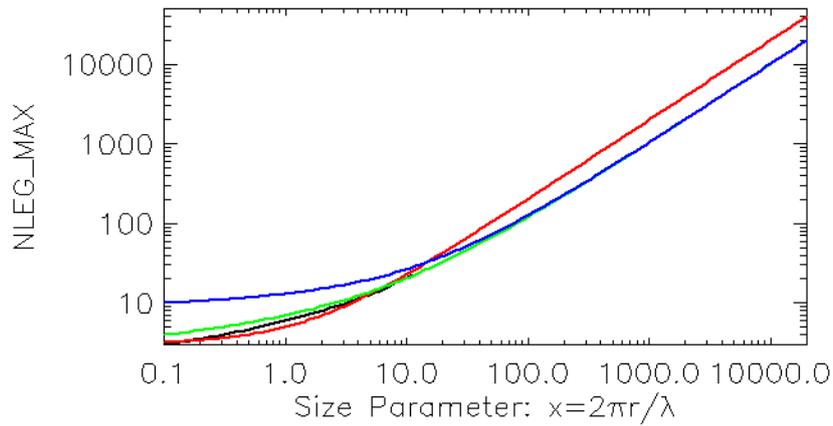
292 Reduction on the number of tabulated wavelengths was based on a two-step procedure. Firstly, we
293 started from the shortest wavelength value in the range of interest, then we searched for the next
294 wavelength value at which any of the two components of the complex refractive index differs more
295 than 1%. This procedure is effective in reducing the number of wavelengths far from the resonances
296 where the spectral variability of the refractive index is large. However, within the spectral range of
297 resonances the above criterion is not so efficient and, hence, a second criterion has been adopted to
298 further reduce the number of wavelengths. This test is similar to the first one (a threshold of 0.1% is
299 adopted this time) except that for the SSOP is based on the differences, computed over the whole
300 set of radii, between extinction coefficients and single scattering albedo values at different tabulated
301 wavelengths. Note that, the adopted criterion adopted allows the use of the smaller tabulated
302 wavelength in the computation without need of interpolation.

303 For some species, the wavelength range for which tabulated values of the complex refractive index
304 are available does not cover the overall range of wavelengths of interest considered in this study.
305 No attempt was done to extend the original range for example with extrapolation of the values
306 through a functional fit. We discarded this possibility because of the high level of uncertainty in the
307 extrapolation process and the high sensitivity to the fitting procedure adopted (functional form and
308 fitted dataset).

309 Compared to the most common way to implement LUTs for aerosols scattering, we stored single
310 particle optical properties rather than values computed for polydispersions for which a distribution
311 of sizes is taken into account (e.g. Hess et al. 1998). This has been done in order to allow the
312 computation for any possible size distribution. From the point of view of inversion algorithms, this
313 choice results in more flexibility for choice of retrievable quantities, allowing the retrieval of the
314 size distribution itself. A direct consequence of storing single particles SSOPs is the need of a larger
315 number of Legendre coefficients to correctly represent the phase matrix elements. For example,
316 Figure 3 shows the results of 4 different parameterizations available in literature estimating the
317 minimum number of required Legendre coefficients as a function of the size parameter x , including
318 the one adopted in MIEV0. It is evident that large x values (i.e. for large radii and short
319 wavelengths) require more than 10000 Legendre expansion terms. However, these
320 parameterizations do not take into account the value of the complex refractive index. For example,
321 large values of the imaginary part result in a smoother phase function and, therefore, a smaller
322 number of Legendre coefficients is required.

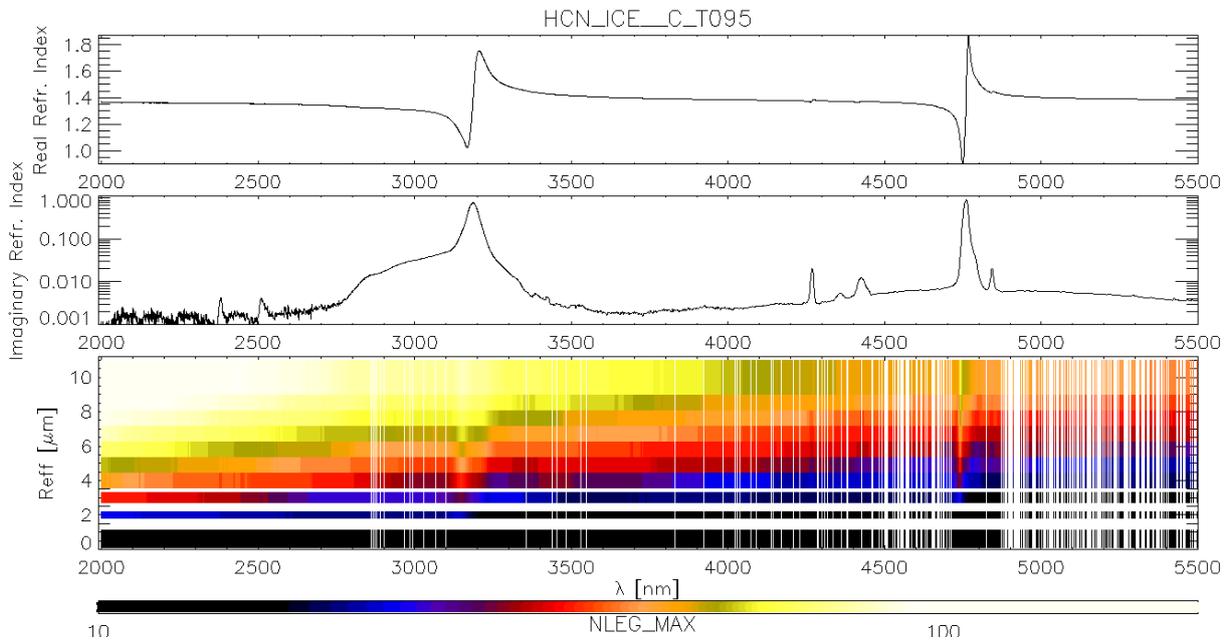
323 The number of Legendre coefficients can be truncated with methods like the δ -M (Wiscombe,
324 1977) or others (Nakajima and Tanaka 1988; Hu et al. 2000; Mitrescu and Stephens 2004; Lin et al.,
325 2018; for a review see Sharma 2015) that, however, are usually applied to integrated
326 polydispersions. For example, in the widely used DISORT2 solver (Stamnes et al., 2000) the δ -M is
327 automatically adopted consistently with the input characteristics of the required simulation.

328 Since for the above reasons, these methods are not suitable for our LUTs, we perform an
329 optimization on the number of Legendre coefficients based on the phase functions reconstruction
330 for polydispersions, for which less coefficients are required.



331

332 **Figure 3: Minimum number of Legendre coefficients required to describe the phase function of a**
 333 **single homogeneous spherical particle of HCN ice, computed according to Wiscombe (1980) (Black),**
 334 **Lenoble (1993) (Red), Bohren and Huffman (1983) (Green) and Mishchenko et al (2002) (Blue).**



335

336 **Figure 4: Top and middle panels: real and imaginary parts of HCN ice refractive index. Lower panel:**
 337 **Minimum number of Legendre coefficients required to describe the phase function of a lognormal size**
 338 **distribution of spherical particles with $v_{\text{eff}} = 0.3$ computed according to Wiscombe (1980). Blank**
 339 **regions represent wavelengths and radii removed after the optimization process (see Section 3.2).**

340

341 There is no general parameterization for the choice of the number of Legendre coefficients for
 342 polydispersions. For example, Ding et al. (2009) found that a maximum of 32 Legendre coefficients
 343 are required to represent ice and water clouds (16 for dust aerosols). But, besides the fact that this
 344 low number is obtained after applying the δ -fit method (Hu et al, 2000) to the original SSOPs, they
 345 found that truncated phase functions cannot match perfectly the original counterparts at some
 346 scattering angles. This is evident, for example, in the rainbow region for a water cloud and sharply
 347 peaked halos for an ice cloud. On the other hand, Kokhanosky et al (2010) used up to 720 Legendre
 348 expansion terms, while Rozanov and Lyapustin (2010) found a value of 680 depending on the code,
 349 the wavelength and the microphysics of the polydispersion (water cloud vs aerosols). An example

350 of the dependence of the minimum number of Legendre coefficients with the particles effective
351 radius (r_{eff}) and wavelength for a lognormal size distribution (Hansen and Trevis, 1974) of HCN
352 particles is given in Figure 4.

353 For the above reasons, a species- and size distribution-dependent procedure to define the minimum
354 number of coefficients was developed to optimize the LUTs. The optimization procedure was based
355 on the comparison of the computed phase functions for a set of size distributions. We evaluated that
356 Legendre polynomials of orders greater than 5000 contribute less than 10^{-3} to the phase function
357 reconstruction for all the species and wavelengths in Table 3. Then we iteratively decreased the
358 number of Legendre coefficients until the average percentage difference between the initial and the
359 current phase function, computed over 181 regularly spaced scattering angles in the entire range
360 (0° - 180°), is larger than 1%.

361 For the above optimization procedure, the log-normal functional form has been assumed as
362 size distribution, being one of the most commonly adopted to perform RT studies of planetary
363 atmospheres. This distribution is parameterized with the particles modal radius (r_g) and standard
364 deviation (σ_g) (Hansen and Trevis, 1974). We computed r_g and σ_g from a discretized set of r_{eff} and
365 effective variances (v_{eff}) (Hansen and Trevis, 1974), since these are the quantities that are usually
366 compared in RT studies of planetary atmospheres. The ranges of effective radii considered to
367 generate the size distributions depend from the species and their value is reported in Table 4. Such
368 ranges are defined to ensure that the LUT is suitable to study the different atmospheres of the Solar
369 System planets. To generate a set of size distributions the overall r_{eff} range (0.01-100 μm) was
370 discretized by defining 10 equally spaced values for each order of magnitude for a total of 37
371 values. For the effective variances (v_{eff}) we considered for all species the values $v_{eff} = 0.1, 0.2, 0.3,$
372 0.4 and $0.5 \mu\text{m}$, ranging from the minimum to the maximum values found in literature (Ramirez-
373 Beltran et al., 2009; Karkoschka & Tomasko, 2005; Roman et al., 2013; Millour et al., 2015). With
374 the above assumptions for a given species, up to 185 size distributions are used in the Legendre
375 coefficients optimization. Note that a similar approach also includes unrealistic size distributions
376 such as the one with very large r_{eff} and small v_{eff} . More realistic size distribution are probably
377 obtained with effective variance values proportional to the effective radius one (e.g. Gavrilova &
378 Ivlev 1996). However, as expected these size distributions characterized by large r_{eff} and relatively
379 small v_{eff} are the ones that require the largest number of coefficients. Therefore, even if probably
380 less representative of real polydispersions in planetary atmospheres, the set generated was used to
381 obtain a conservative estimation of the number of Legendre coefficients required to reliably
382 represent the phase functions (see Table 4). The obtained values range roughly from 100 to about
383 4600. As expected, the latter value is needed for wavelengths in the UV range for polydispersions
384 with a large r_{eff} and intermediate v_{eff} and for species with relatively high real component of the
385 refractive index and corresponding low value for the imaginary part (Figure 4).

386 Regarding the use of a large number of Legendre coefficients in numerical simulations, it
387 should be noted that, in previous versions of multiple scattering RT solvers (e.g. Stamnes et al
388 1988, Evans and Stephens 1991) the number of Legendre coefficients was proportional to the
389 number of streams for the simulation, increasing drastically the computational needs. However, this
390 is no longer valid for recent solvers (e.g. Lin et al. 2015) as well as for Monte Carlo based ones.

391

392 **4. Computational time improvement and LUTs application to planetary data**

393

394 The improvement in terms of computational time for using the LUTs with respect to the dynamic
395 computation of aerosols and gases properties every time they are needed has been tested on a PC

396 equipped with an Intel Core i5 CPU650 with a 3.2 GHz frequency and 3.8 Gb of system RAM. The
397 real-time computation of the scattering properties of a polydispersion of H₂O ice at 80 K in the
398 wavelength range 1111 nm – 5500 nm considering more than 3000 spectral points (the procedure
399 adopted to resize the spectral grids is described in Section 3.2), is performed in about 80 minutes
400 while just a few seconds are required to read the SSOPs LUTs. Similarly, the computation of the
401 absorption coefficients of CH₄ considering the wavelength range and resolutions provided in Table
402 2 requires 14 minutes for a single set of P,T and χ (i.e. for a single atmospheric layer). On the other
403 hand, the reconstruction of the same absorption coefficients using the GACs LUTs is performed in
404 less than 1 minute.

405 For the reasons above, the advantage in using the LUTs with respect to the real-time computation is
406 evident. The application of the LUTs to RT computations has been tested for case studies on Saturn,
407 Mars and Jupiter.

408 In the case of Saturn, we exploited the GAC LUTs for CH₄ to model VIMS data in the 1.5 – 4.0 μm
409 wavelength range observing a faint luminescent ring in the northern polar cap of the planet (Adriani
410 et al., 2015).

411 Jupiter data in the range 2.4 – 3.0 μm from the JIRAM spectrometer have been analyzed taking
412 advantage of CH₄ and H₂O GAC LUTs, and tholin and NH₃ SSOPs LUTs (Sindoni et al., 2017).

413 CO₂ and H₂O GAC LUTs have been used together with the Mars dust SSOP LUT to study a
414 Martian local storm in the range 0.5 – 2.5 μm observed by the OMEGA spectrometer (Oliva et al.,
415 2018). Moreover, the same LUTs are being exploited to study OMEGA limb data in the same
416 wavelength range.

417 **5. Summary and future developments**

418 We have described a database for gaseous and aerosols optical properties in the VIS-NIR
419 spectral range (400-5500 nm) for some of the main components of planetary atmospheres. We
420 focused on CH₄, CO₂, and H₂O molecular species for gaseous absorption, and on the scattering
421 properties of condensates likely forming the bulk of atmospheric particulates (Table 4). This
422 database is valuable as input for RT calculations especially for the atmospheres of Venus, Mars,
423 Jupiter, Saturn, and Titan. Gaseous coefficients have been computed with the LBL approach
424 adopting Voigt line shapes for several combinations of pressures, temperatures and mixing ratios
425 suitable to describe the atmospheres of rocky and gaseous planetary environment. A new
426 interpolation method tested on these LUTs is suggested for obtaining absorption coefficients for the
427 desired values of P, T and χ with improved accuracy.

428 On the other hand, for the SSOPs of the species of interest, we assumed homogeneous spheres
429 to compute the LUTs for single particles. Mie theory has been adopted within the limit $0.1 < x <$
430 20000 and Rayleigh for particles with $x < 0.1$. Within the LUTs we provide the Legendre
431 coefficients of phase functions, in a number sufficient to accurately describe the considered radii
432 polydispersions within the ranges given in Table 4.

433 The LUTs discussed here must be thought as forming a first version of the database. Several
434 developments are foreseen in the next future. The gaseous line parameters will be extended to
435 include the recently distributed HITRAN 2016 version (Gordon et al., 2017). Further molecules,
436 such as CO, NH₃, PH₃, that are present in many atmospheres as trace gases, will be processed and
437 inserted in the database. Moreover, we will evaluate the possibility to produce LUTs with different
438 line shapes/broadenings and we will extend temperature, pressure and mixing ratio ranges to
439 account for the most extreme environments in the Solar System.

440 We plan analogous upgrades for the SSOPs LUTs, with the inclusion of other species, like
441 hydrocarbon ices, polycyclic aromatic hydrocarbons (PAH) and mixtures. The inclusion of non-
442 spherical and coated particles, expected to be present in the outer Solar System, will be evaluated to
443 account for different microphysical regimes.

444

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446

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450

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