LOTOS-EUROS Technical Report

LETR-2016-001

Aerosol modes

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1.1   Diameter counts

The aerosol composition in the volume is usually described by a size distribution over several separate size modes, see e.g. Seinfeld and Pandis 2006, Chapter 8. Each size mode collects all particles in a certain size range. Which sizes are present in a collection of aerosols depends on their composition and on the the physical processes acting on them.

To describe the aerosols in a size mode, the first action would be to measure the sizes. First a number of size bins is defined for the aerosol diameters. A size bin i is centered around a diameter D_i and has a width ΔD_i. Each of the aerosols is assigned to one of the bins, and the number of aerosols per bin is denoted by:

$$\Delta N_i \;=\; |\{\text{aerosols in bin around } D_i\}| \tag{1.1}$$

The upper panel of Figure 1-1 shows a a typical example of how ΔN_i could look like as function of the diameter D_i. The total number of aerosol particles in this example is 10^6.

The number of aerosols that is present in a size bin depends on the total amount of aerosols present and on the width of a bin. In the current example all bins have the same width, but if the bins were defined differently, the shape of figure would have have been different as well. To not let the aerosol characterization depend on the choice of the bins, it is better to use the *number density*, which divides the number of particles by the bin size and the total number:

$$\frac{1}{N}\,\frac{\Delta N_i}{\Delta D_i} \tag{1.2}$$

The number density is plotted in the middle panel of Figure 1-1. The area in the bins is now equal to 1.

Since aerosols diameters could differ several orders of magnitude, it is common practice to not use diameters D but the logarithms $\ln D$. If the diameter bins are defined around regular spaced values of $\ln D$, the number densities turn out to show a normal distribution as seen in the lower panel of Figure 1-1. Thus, the logarithms of the diameters seem distributed according to a normal distribution, which is characterized by a mean and variance:

$$\ln D \;\sim\; \mathcal{N}\left(\,\mu\,,\,\sigma^2\,\right) \tag{1.3}$$

An equivalent definition is to say that the diameter D has a *log-normal* distribution. For log-normal distributions it is common practice to characterize it by the *geometric mean* and *geometric standard deviation*:

$$D_g \;=\; e^{\mu} \qquad \Leftrightarrow \qquad \mu \;=\; \ln D_g \tag{1.4}$$

$$\sigma_g \;=\; e^{\sigma} \qquad \Leftrightarrow \qquad \sigma \;=\; \ln \sigma_g \tag{1.5}$$

To characterize an aerosol sample it is therefore 'sufficient' to measure all the log-diameters, compute the mean μ and standard deviation σ over the population, and convert this to geometric mean D_g and geometric standard deviation σ_g.

1.2   Properties of log-normal distributions

The probability density function represents the number density for infite small size bins (dashed line in the lower panel of Figure 1-1). For the log-diameter and 'ordinary' diameter the probability distributions are:

$$\frac{\mathrm{d}N}{\mathrm{d}\ln D} \;=\; f(\ln D; \ln D_g, \ln^2 \sigma_g) \tag{1.6a}$$

$$\frac{\mathrm{d}N}{\mathrm{d}D} \;=\; \frac{\mathrm{d}N}{\mathrm{d}\ln D}\frac{\mathrm{d}\ln D}{\mathrm{d}D} \;=\; f(\ln D; \ln D_g, \ln^2 \sigma_g)\; D^{-1} \tag{1.6b}$$
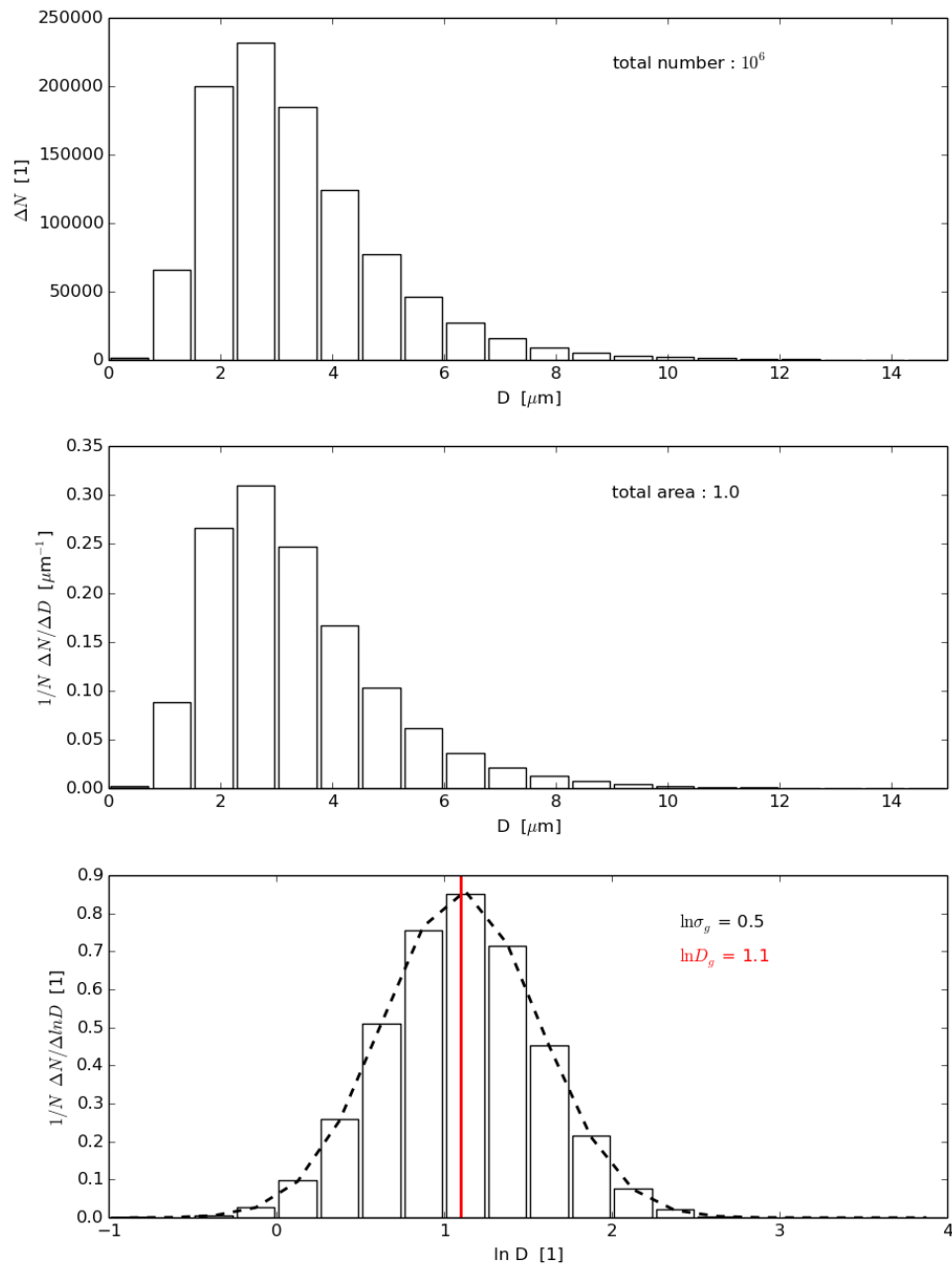
Figure 1-1   Aerosol number and number-densities of randomly generated aerosol population.

where:

$$f(x; \mu, \sigma^2) \;=\; \frac{1}{\sqrt{2\pi\sigma^2}} e^{-\frac{(x-\mu)^2}{2\sigma^2}} \tag{1.7}$$

A number of special diameters could be computed given the distribution. We will use abbreviations 'g' for geometric mean and standard deviations, 'm' for mode, and 'a' for arithmetic mean.

A lognormal distribution (in practice termed *mode*) is characterized by the (diameter) value where the probability density peaks. It is related to the geometric mean by:

$$D_m \;=\; D_g \, e^{-\ln^2 \sigma_g} \tag{1.8}$$

The *median* of a log-normal distribution is is the diameter for which half of the particles is smaller and half of the particles is larger. It is equal to the *geometric mean*. It is advised to always use the name geometric mean, since this together with the geometric standard deviation defines the distribution. By lack of a 'median' standard deviation, it is not useful to use a 'median' diameter in this context.

For particles it is imporant to know properties of the moments D^γ, which are related to diameter ($\gamma = 1$), surface and cross-section ($\gamma = 2$), and volume ($\gamma = 3$). A property of the log-normal number distribution is that the moments also have a log-normal distribution within the population. The *geometric mean* of a moment γ could be computed from the number geometric mean:

$$D_{g\gamma} \;=\; GM[D^\gamma] \;=\; D_g \, e^{\gamma \ln^2 \sigma_g} \tag{1.9}$$

Combining (1.8) and (1.9) gives the following equation for the mode of a moment:

$$D_{m\gamma} \;=\; D_g \, e^{(\gamma-1)\,\ln^2 \sigma_g} \tag{1.10}$$

For the *arithmetic mean* of a moment the following relation holds:

$$E[D^\gamma] \;=\; e^{\gamma \ln D_g + 1/2\gamma^2 \ln^2 \sigma_g} \;=\; \left(D_g \, e^{\gamma/2 \, \ln^2 \sigma_g} \right)^\gamma \tag{1.11}$$

Thus, the expected value of a moment is equal to a moment of a special diameter, which represents an average diameter that would represent the average moment if only one diameter was present in the total population:

$$D_{a\gamma} \;=\; (E[D^\gamma])^{-\gamma} \;=\; D_g \, e^{\gamma/2 \, \ln^2 \sigma_g} \tag{1.12}$$

For each of the relevant moments, some relevant properties and features are discussed in the following sections. The various formula available have been tested for the sample distribution, and the results are shown in Figure 1-2 as function of D and Figure 1-3 as function of $\ln D$. For these plots the number of bins has been increased, and instead of using bars, the densities are denoted by gray dots. Densitiy functions are added according to eq. 1.6, and should be in agreement with the sampled number densities.

1.3   Moment 0 : number

The zero moment represents the particle numbers. The top panels of figures 1-2 and 1-3 show the number densities of the example aerosol population. The area under the denisty is exactly 1.

For the density versus diameter, the only useful derived quantitiy is the mode (green line), which indeed aligns with the peak in the distribution.
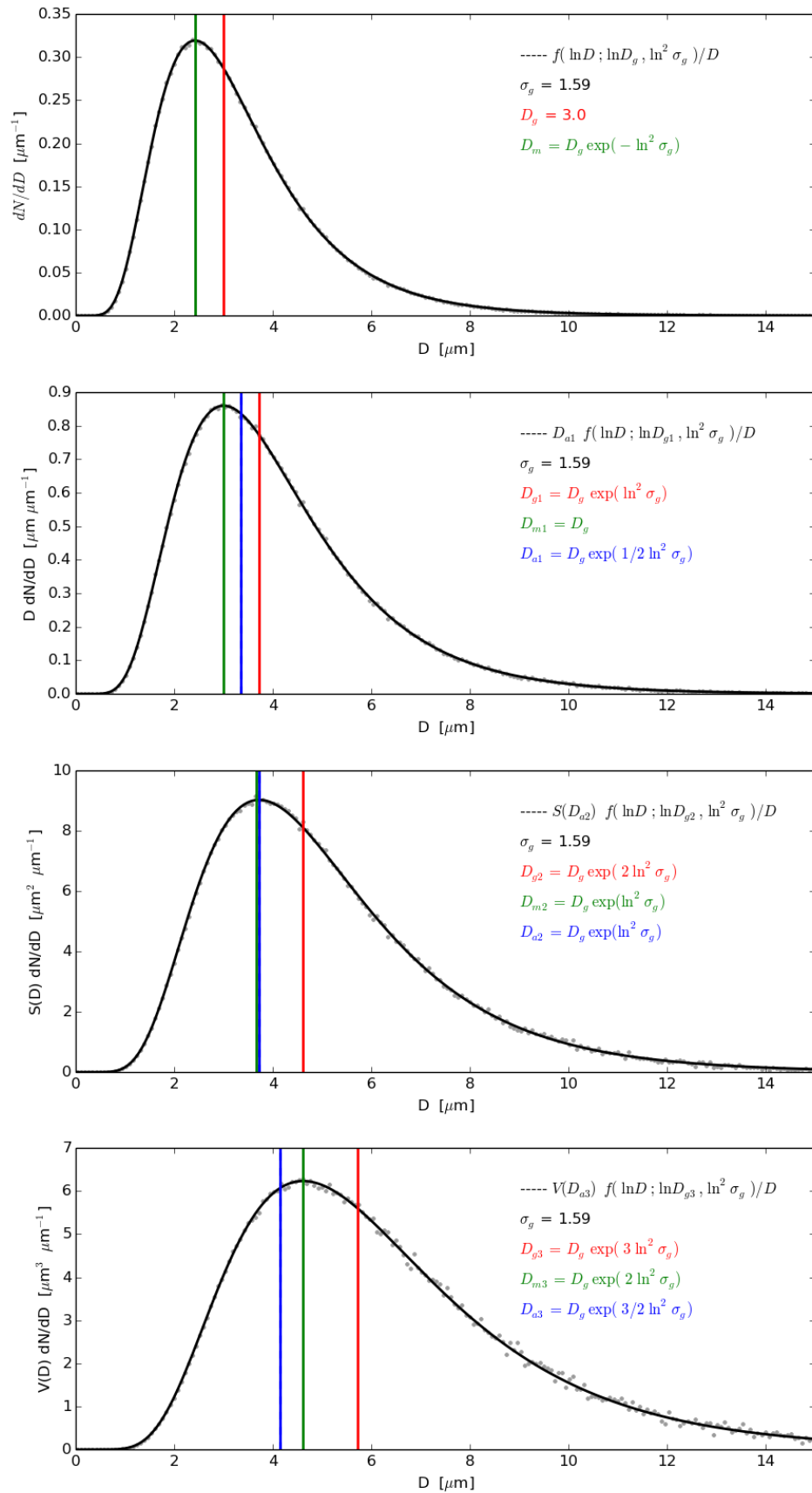
Figure 1-2   Aerosol number, diameter, surface, and volume densities versus diameter for randomly generated aerosol population.
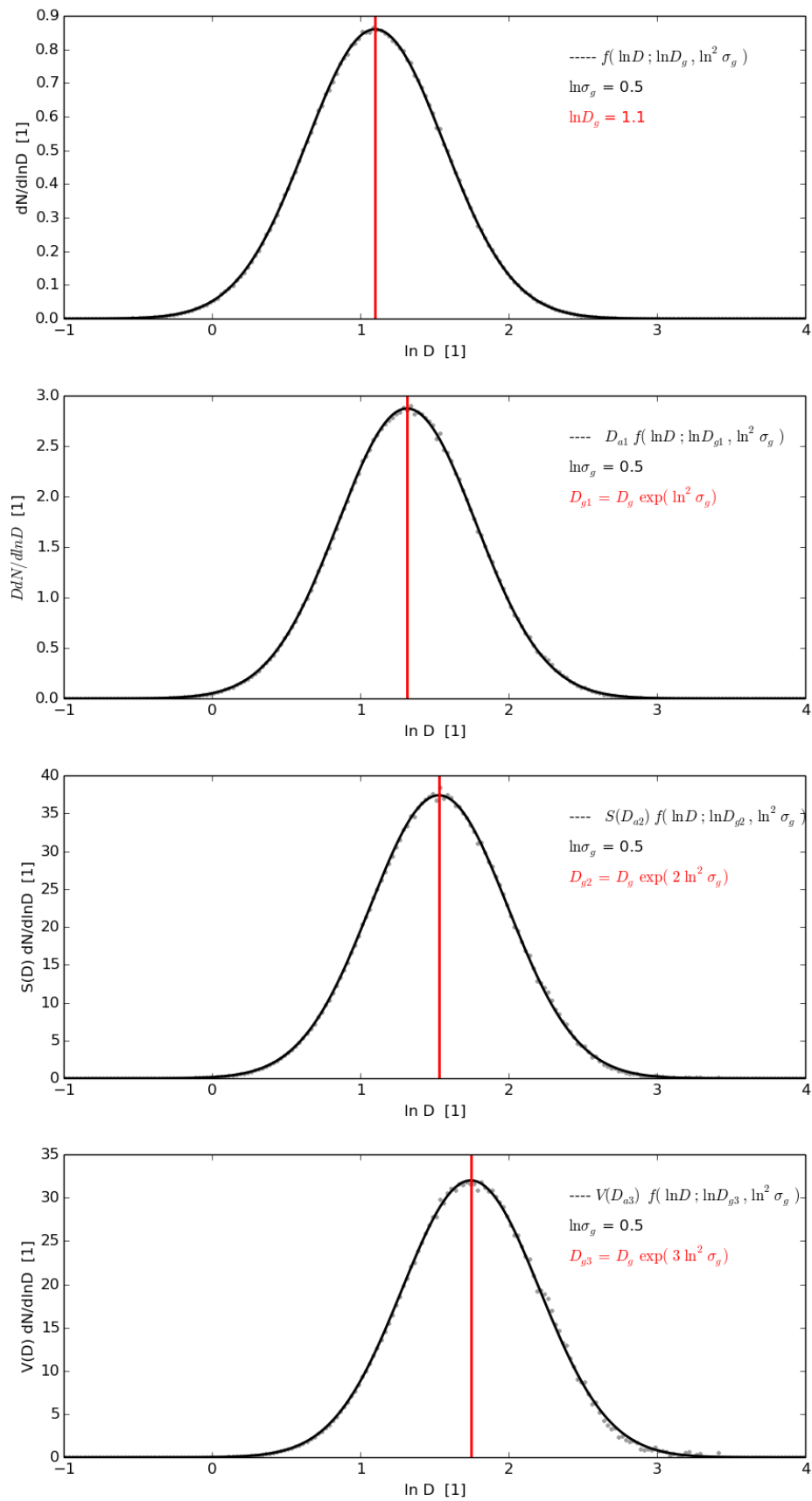
Figure 1-3   Aerosol number, diameter, surface, and volume densities versus log-diameter for randomly generated aerosol population.

1.4   Moment 1 : diameter

The first moment is related to the diameters in the population. The *surface density* desribes the relative amount of diameter present in aerosols with a certain diameter:

$$\frac{\mathrm{d}N_D}{\mathrm{d}D} \; = \; D\,\frac{\mathrm{d}N}{\mathrm{d}D} \tag{1.13}$$

Since the larger particles contribute more to the diameter than the smaller, the peak in the diameter density i shifted to the right compared to the number densities (second panels in figures 1-2 and 1-3). The area under the curve is equal to the average diameter.

The *geometric mean diameter* is computed from (1.9) using $\gamma = 1$:

$$D_{g1} \; = \; D_g\, e^{\ln^2 \sigma_g} \tag{1.14}$$

The *mode diameter* defines the size which occures most in the distribution, and is equal to the number geometric mean following eq. (1.10) for $\gamma = 1$:

$$D_{m1} \; = \; D_g \tag{1.15}$$

The green line in the second panel of Figure 1-2 shows that it is indeed aligned with the maximum proability.

The *arithmetic mean diameter* defines the average diameter of the population. Following eq. (1.12) this could be computed from:

$$D_{a1} \; = \; D_g\, e^{1/2\ \ln^2 \sigma_g} \tag{1.16}$$

The blue line in the second panel of Figure 1-2 denotes the computed arithmetic mean, and it has been been verified that this is similar to the sample mean computed over the population.

1.5   Moment 2 : cross-section and surface

For a spheric aerosols, the cross-section and surface could be computed from the diameter following:

$$
\begin{aligned}
C(D) &= \pi\,(D/2)^2 = \tfrac{1}{4}\pi\,D^2 & \text{(1.17a)}\\
S(D) &= 4\pi\,(D/2)^2 = \pi\,D^2 & \text{(1.17b)}
\end{aligned}
$$

Since these quantities only differ a factor, the derived diameters are the same; for simplicity, we only show results for the surface values.

The *surface density* desribes the relative amount of surface present in aerosols with a certain diameter:

$$\frac{\mathrm{d}N_S}{\mathrm{d}D} \; = \; S(D)\,\frac{\mathrm{d}N}{\mathrm{d}D} \tag{1.18}$$

The *surface geometric mean diameter* is computed from (1.9) using $\gamma = 2$:

$$D_{g2} \; = \; D_g\, e^{2\ \ln^2 \sigma_g} \tag{1.19}$$

This geometric mean defines the position of the density curve. The area under the curve is equal to the average diameter.

The *surface mode diameter* defines the size which contributes most to the total sufrace present in the population, and could be computed from the number geometric mean following eq. (1.10) for $\gamma = 2$:

$$D_{m2} \; = \; D_g\, e^{\ln^2 \sigma_g} \tag{1.20}$$

The green line in the panel shows that it is indeed aligned with the maximum proability.

The *surface arithmetic mean diameter* defines an average diameter that could be used to compute the total amount of surface present. Following eq. (1.12) this could be computed from:

$$D_{a2} \;=\; D_g \, e^{\ln^2 \sigma_g} \tag{1.21}$$

The blue line in the panel of Figure 1-2 denotes the computed value, and it has been been varified that this is similar to the diameter obtained from the sample mean surface of the population.

1.6   Moment 3 : volume

For a spheric aerosols, the volume could be computed from the diameter following:

$$V(D) \;=\; \tfrac{4}{3}\pi \, (D/2)^3 \;=\; \tfrac{1}{6}\pi \, D^3 \tag{1.22}$$

The *volume density* desribes the relative amount of surface present in aerosols with a certain diameter:

$$\frac{\mathrm{d}N_V}{\mathrm{d}D} \;=\; V(D) \, \frac{\mathrm{d}N}{\mathrm{d}D} \tag{1.23}$$

The *volume geometric mean diameter* is computed from (1.9) using $\gamma = 3$:

$$D_{g3} \;=\; D_g \, e^{3 \, \ln^2 \sigma_g} \tag{1.24}$$

This geometric mean defines the position of the density curve. The area under the curve is equal to the average volume.

The *volume mode diameter* defines the size which contributes most to the total volume present in the population, and could be computed from the number geometric mean following eq. (1.10) for $\gamma = 3$:

$$D_{m3} \;=\; D_g \, e^{2 \, \ln^2 \sigma_g} \tag{1.25}$$

The green line in the panel shows that it is indeed aligned with the maximum proability.

The *volume arithmetic mean diameter* defines an average diameter that could be used to compute the total amount of volume present. Following eq. (1.12) this could be computed from:

$$D_{a3} \;=\; D_g \, e^{3/2 \, \ln^2 \sigma_g} \tag{1.26}$$

The blue line in the lower panel of Figure 1-2 denotes the computed value, and it has been been varified that this is similar to the diameter obtained from the sample mean volume of the population.

2   Aerosol size in LOTOS-EUROS

2.1   Summary

Aerosol concentrations can be divided over size bins or desrcibed as modes.For atmospheric aerosol often Nucleation, Aitken, accumulation and coarse mode are discerned.One has to be careful: D_{g3} for the mass distribution of the Aitken mode is different from D_g1 for the number distribution. Which diameter to to take depends on the process. Therefore a unique diameter should be defined to which all related diameters are related. The impact of processes on the diameter (heavy particles fall out more quickly so the mass mean/median diameter of the coarse mode becomes smaller) is neglected in the current approach in LOTOS-EUROS, which is acceptable given other uncertainties.

LOTOS-EUROS is designed to model mass of particles in the diameter size bins 0-2.5 μm and 2.5-10 μm since these are observed quantities and used for regulations. In these observations, the particle sizes are including absorbed water. For some aerosols, like sea spray, the corresponding dry diameter would be a factor two smaller, assuming a relative humidity of 80%. In reality the size changes with RH, but this is nog taken into account in this crude mass-based bin definition. It is reflected in the assumed particle density per specie. The two bins are assumed to contain the mass in the accumulation mode and coarse mode, with mass mode diameter $D_{m3}$of 0.7 μm and 8 μm respectively. These diameters are used to calculate deposition velocities.

Emissions are traditionally described and reported in terms of mass, and are attributed to the accumulation and coarse mode, for LOTOS-EUROS to the fine and coarse size bins. The antrhopogenic emissions are described in more detail in the next section and in the LOTOS-EUROS reference guide. For the emission of sea spray and desert dust, emission functions are explicitly size dependent and we have more detailed information about the size distribution. Given the high concentrations in emission areas, the wide distribution and the strong size-dependency of deposition, for these species it is worthwhile to specify the deposition velocity in more than 2 size bins, and the fine and coarse size bin are divided in 2 respectively 3 size bins. For other species, just one fine and one coarse size bin is assumed.

The basic definitions found in code are listed in table 2.1 and table 2.2. These are in fact mass-based bins that can be grouped in modes: ff + f together represent single fine (accumulation) mode, ccc+cc+c represent single coarse mode. So we should define a single σ and D_g per mode. The basic assumption in this process is that the diameters used for the calculation of deposition velocities are wet diameters representing the D_{m3} so that the dry diameters are half of this,except for dust which hardly takes up water.

accumulation, D_{m3}=0.7μm, σ=1.59					
D_g	D_m	D_{ag}	0.46	0.37	0.46
D_{g1}	D_{m1}	D_{a1}	0.56	0.45	0.51
D_{g2}	D_{m2}	D_{a2}	0.70	0.56	0.56
D_{g3}	D_{m3}	D_{a3}	0.87	0.70	0.63
coarse, $D_{m3} = 8\mu m$, σ=2.0					
D_g	D_m	D_{ag}	3.06	1.89	3.06
D_{g1}	D_{m1}	D_{a1}	4.95	3.06	3.89
D_{g2}	D_{m2}	D_{a2}	8.00	4.95	4.95
D_{g3}	D_{m3}	D_{a3}	12.93	8.00	6.92

Table 2.1   Diameter values of various relevant mode characteristics. Diameters in μm. For dry particles except dust these diameters should be halved

2.2 Emissions

Emission inventories are based on officially reported emission masses and estimates of mass emissions for other sectors. LOTOS-EUROS reads in these mass emissions and distributes them over the species according to distribution tables. Particulate matter emissions databases are defined in mass (kg, ton) per size bin (fine and coarse, (0-2.5μm and 2.5-10μm). Examples of such emission inventories are TNO-MACC emissions (Kuenen et al. 2014) and EDGAR emissions.

A special set of particle number emissions is also available at TNO. Information on geometric mean, andstandard deviations is available per emission source, with estimates of the mass contribution of EC, OC, SO4 and mineral components Since most of the particles are in the Aitken and accumulation modes, this inventory considers only the size range 0.01-0.3μm. This emission inventory describes the number distribution better than results based on a mass-based emission inventory but misses an important part of the mass, which is mainly contained in the larger particles. Note that in short time after emission (minutes to few hours), the smallest particles are coagulated with larger particles and contribute to mass but no longer to number, or have grown to larger particles by condensation of co-emitted gases. One has to check whether this has been taken into account in the emission inventory or should be taken in to account by using a preprocessor, since LOTOS-EUROS does not resolve these scales. The use of this emission inventory is described in Kukkonen et al. 2016

2.3 Deposition and sedimentation

One value of the deposition velicity is caluclated per size bin, thus accounting for the size dependency of the process. In the deposition process, the same mass-based radius and density is used for all species within the size bin. This density is not the density of the dry species but an average of a wet (and internally mixed) particle. For dust this would imply a too small density, for other species the assumption would be reasonable for 80% RH.

$$\rho = 1.14e3 kg/m^3 \tag{2.1}$$

process	**bin**	1	2	3	4	5
	name	ff	f	ccc	cc	c
binsize		0.14-1	1-2.5	2.5-4	4-7	7-10
emissions input	size range		[0,2.5]			[2.5,10]
online sea spray	size range	0.14-1	1-2.5	2.5-4	4-7	7-10
online dust	size range	0.14-1	1-2.5	2.5-4	4-7	7-10
diameter for deposition	related to mass/volume in bin	0.33	0.70	3.00	5.00	8.00
`le_partsize`						

	mode	Accumulation	Coarse
optics	D_g dust	0.45	3.06
optics	D_g other	0.23	1.50
	based on	D_{m3}=0.7	D_{m3}=8
optics	surface mean diameter dust	0.56	4.9
optics	surface mean dry diameter other	0.28	2.47
`le_radiation`	σ_g	1.59	2
optics	number mean diameter dust	0.37	1.89
optics	number mean dry diameter other	0.18	0.94
`le_radiation`	σ_g	1.59	2

Table 2.2   Definitions of modes and bins. Diameters in μm.

Gravitational settling (sedimentation) is only relevant for coarse particles. For size bins 2.5-7 μm the contribution of gravitational settling to deposition is calculated but has a small effect and could be left out.

2.4 Heterogeneous chemistry

Reactions between gas and water phase in water shell around aerosols are accounted for in le_chem_hetro.F90 Three aerosol distributions are used:

- For sulphate/SIA related reaction (NH4SO4a_f),we take aerosol size from Whitby accumulation mode (1978) Number mean dry radius: 0.034um, sigma=2

- fine mode sea salt (Jaenicke R., 1993, Ed. Hobbs).
  Number mean radius = 0.1 um; sigma=1.9

- coarse mode sea salt (Haldis 2009,).
  Number mean radius = 1 um; sigma=2.0

The code mentions that the size distribution NH4SO4a_f is for the accumulation mode, but the present number suggests that it is the Aitken mode.

For SIA, aerosol water is taken from Isorropia II, which takes into account the water that is attached to the aerosol including NH4NO3. This is similar to the approach by Gerber to determine the growth of aerosol using hygroscopicity as applied in the radiation routine. But the difference is that in Isorropia II severeal of these growth functions are combined.

For sea salt, a wetness factor is determined explicitly to determine the wet radius. To calculate the number of aerosol, the wet density $\rho = 1.14e3 kg/m^3$ is used the radius is assumed to be 'dry radius. When determining radii of the particles, for sea salt only concentrations in c and f bins are taken into account, not ff and ccc and cc in le_chem.F90 and le_chem_hetro.F90, thus assuming two modes, but the mass in the cc, ccc and ff size bins is taken into account in determining the reactions.

2.5 Boundary conditions from other models

LOTOS-EUROS uses boundary conditions from CIFS and other CAMS products. Based on their internal approach (modes or size bins) CIFS delivers dust and sea spray mass in several size bins. Based on the consideration that CIFS classifies their size bins in terms of dry radius we come to the following distribution of CIFS aerosol modes to LOTOS-EUROS size bins. It should be noted that currently, the boundary conditions for sea spray from CIFS/MACC are not used by the model, as they lead to an overestimation of the concentrations.

CIFS name	dry size μm	Na_ff	Na_f	Na_ccc	Na_cc	Na_c
Aermr01	0.03-0.5	100%	-	-	0	-
Aermr02	0.5-5	-	10%	20%	40%	30%
Aermr03	5-20	-	-	-	-	-
CIFS name	**dry size μm**	Dust_ff	Dust_f	Dust_ccc	Dust_cc	Dust_c
Aermr04	0.03-0.55	2%	8%	10%	40%	40%
Aermr05	0.55-0.9	2%	8%	10%	40%	40%
Aermr06	0.9-20	2%	8%	10%	40%	40%

Table 2.3   Translation of CIFS dust and sea spray to LOTOS-EUROS size bins

2.6   Optical properties

2.6.1   Size modes

The base version of LOTOS-EUROS was able to calculate AOD values based on a mass-based extinction coefficient. Values were representative for 550nm, the most common wave length for many applications in the past. This routine is still present but is not able to calculate the optical properties that are typically needed for a radiative transfer code, like extinction coefficient, single scattering albedo and asymmetry parameter. A radiative transfer code can be used for several applications, e.g. a two-way coupling to a meteorological model like RACMO2, or to calculate the impact of emission scenarios on incoming solar radiation for the solar spectrum. The extinction coefficient by itself is useful to estimate the impact of aerosol on visibility. Present-day earth observations from AERONET or satellites like POLDER cover not only AOD but also microphysical properties are retrieved. These microphysical properties are underlying the calculation of AOD and other paramters. To fully exploit the possibilities of LOTOS-EUROS a more advanced scheme was implemented. The approach is described here.

Optical Properties per a particle are determined using an effective medium approach. The effective medium is an imaginary material that represents the optical properties of the mixture of which the aerosol particle is composed. The refractive index of the effective medium can be calculated from the refractive indices and volume fractions of the components of the real aerosol. Mie-scattering calculations demand a significant computational burden and simplifying the Mie-scattering theory causes significant errors. To tackle this problem, a lookup table is pre-calculated for dimensionless aerosol sizes. The input parameters of this lookup table (refractive index and size) are sampled with forty times fifteen values for the refractive index (real Œ imaginary) and a hundred values for the size parameter. With interpolation, the discretization error is expected to be low (at most a few percent). In this way, the AOD calculations can be made in an accurate and at the same time a cost-efficient way.

The cross section of aerosol, together with its refractive index, are the key elements to obtain their impact on radiation. The modelled aerosols are treated as internally mixed particles per size mode. Four modes are used: two fine modes (dust and non-dust) and two coarse modes (dust and non-dust).Lognormal distributions for the accumulation mode and coarse mode for the mass-based distribution and translate them to distributions of cross section and number by using the relations in Chapter 1. The total mass in a size bin has to be divided over a representative number of particles with a representative size. Per mode, a median dry diameter and standard deviation are defined to translate modelled mass to single particle properties and particle number concentrations, for which water uptake is taken into account. Dust and sea spray mass concentrations are divided over more than two size bins. The size binsshould be considered as a coarse subsampling of these modes. For the calculation of optical properties, the mass in the two fine size bins is fully assigned to the fine mode and the mass in the three coares mode bins is fully assigned to the coarse mode aerosol. Also the total values for the complete aerosol population and column are calculated. In the following part we consider n species in a mode and $i = 4$ modes.

2.6.2   Microphysical properties

Properties are calculated per mode (accumulation/coarse). These properties only depend on particle size and composition and not on wave length. In the following the caluclation per mode consisting of n species is written out.

Hygroscopic growth

Hygroscopic growth g as a function of relative humidity RH according to Petters&Kreidenweiss using a simple mixing rule for hygrospcopicity coefficient κ

$$\kappa = \sum_n \epsilon_n \kappa_n \tag{2.2}$$

$$g(RH) = \left(1 + \kappa \frac{RH}{1 - RH}\right)^{\frac{1}{3}} \tag{2.3}$$

This curve goes to infinity for values approaching RH=1, therefore a cutoff is used at RH=0.95.

Aerosol mass to volume

Per mode the volume per specie is determined based on dry mass concentration c and density of specie ρ and the dry volume is summed to obtain the total dry volume

$$V_{dry,i} = \sum_{n,i} c_{n,i}/\rho_{n,i} \tag{2.4}$$

To obtain the water volume the growth factor is used

$$V_{water,i} = V_{dry,i} * (g(RH)^3 - 1) \tag{2.5}$$

$$V_{total,i} = V_{dry,i} + V_{water,i} \tag{2.6}$$

Aerosol number concentration

The total number of aerosols is calculated based on the total aserosol volume and the arithmetic mean particle size D_{a3}

$$N_i = V_{dry,i}/(\frac{1}{6}\pi D_{a3,i}^3) \tag{2.7}$$

Refractive index

Refractive indices of aerosol species and water are taken from literature values: OPAC (Optical Properties of Aerosols and Clouds, Hess et al. 1998) tables are used for SO4, NH4, NO3,EC and sea salt, tables from ECHAM (Kinne et al. 2003) are used for PPM, POM and dust, and the values from Segelstein 1981 are used for water. The real and imaginary part of the refractive index Re and Im per mode (and per wave length) are based on a simple volume weighted average of the refractive index per specie and the contribution of the attached water.

The refractive index per mode is obtained by

$$Re_i = \frac{\sum_n V_{dry,n,i} * Re_n + V_{water,i} * Re_{water}}{V_{total,i}} \tag{2.8}$$

$$Im_i = \frac{\sum_n V_{dry_n,i} * Im_n + V_{water,i} * Im_{water}}{V_{total,i}} \tag{2.9}$$

Effective radius and variance

Per mode, the effective radius $r_{eff,i}$ can be determined.

$$r_{eff,i} = \frac{\int r^2 n(r) r dr}{\int r^2 n(r) dr} = \frac{3}{4}\frac{V_i}{C_i} \tag{2.10}$$

It is related to the volume $V_{total,i}$ and the total wet mean cross section $C = N_T * D_{a2_i} * g(RH)$. For a lognormal distribution it is equal to

$$r_{eff} = D_{g3} \exp(-\frac{1}{2}\ln^2\sigma) r_{eff} = D_g \exp(\frac{5}{2}\ln^2\sigma) \tag{2.11}$$

Also an effective variance $v_e ff$ is defined

$$v_{eff} = \frac{\int (r - r_{eff})^2 r^2 n(r) dr}{r_{eff}^2 \int r^2 n(r) dr} \tag{2.12}$$

which for a lognormal distribution equals

$$v_{eff} = \exp(\ln^2\sigma) - 1 \tag{2.13}$$

2.6.3   Columns and values related to size bins

In the above calculations, the values per internally mixed lognormal mode were calculated and comprises of concentration values per grid cell. To compare with aerosol retrievals (e.g. POLDER, AERONET) the values should be combined to yield a single accumulation (fine) mode and a single coarse mode and total column values. This can be done by following the approach taken above, taking into account the height of the layers. Since the dust mode and the other aerosol are externally mixed, they have to be combined to yield values for the accumulation and the coarse mode. This means that for all the expressions above values per layer and per column can be obtained easily, except for the effective variance which is an integral that is now a sum of two lognormal modes and not easily related to bulk properties. Per layer

$$N_{layer} = \sum_i N_i \Delta h \tag{2.14}$$

and analogous to volume and column values

$$r_{eff_{layer}} = \frac{3}{4}\frac{\sum_i V_{total,i}}{\sum_i C_i N_i} \tag{2.15}$$

For the refractive index again a volume weighted approach is used

$$Re_{layer} = \frac{\sum_i Re_i V_{total,i}}{\sum_i V_{total,i}} \tag{2.16}$$

For column equivalents the layer height must be taken into account in this sum. In retrieving remote sensing data the radius is one of the free parameters, whereas the radius is fixed in LOTOS-EUROS. By using a separate dust mode for LOTOS-EUROS this is partly accounted for, as dust is extremely different from the other aerosol in terms of size distribution and hygroscopicity and is mainly externally mixed. Retrieval products often use the fraction of spherical partices, assuming that dust is non-sperical and other particles are spherical.

$$Spherical fraction = \frac{N_T nondust}{N_T(nondust) + N_T(dust)} \tag{2.17}$$

Optical properties

The refractive indices and number concentrations can be used to calculate the extinction, asymmetry factor and single scattering albedo per grid cell. These are the key input for models that calculate impact of aerosol on radiation, either for climate impact studies, meteorological models, or visibility. Such models require parameters at specific wavelenghts (wave lengths bands). The method applied here uses dimensionless lookup tables, which means that the calculation is efficient and flexible with respect to wave length and particle size. The approach and lookup tables are based on the *Generic Aerosol Optical Toolbox* (Aan de Brugh 2013). Look-up tables were created for two aerosol modes, a fine mode with standard deviation 1.59 and a coarse mode with standard deviation 2, consistent with commonly used size distributions, like the M7 aerosol module that can be used with LOTOS-EUROS.

Optical properties are determined per size class and take into account composition but they also differ per wave length. In LOTOS-EUROS, several common sets of wave length bands and their central line are defined in lotos-euros-radiation.rc. These are

- the 14 shortwave wavelengths that are used by RRTM (used for 2-way coupling with the RACMO2 climate model of KNMI and estimation of the impact of emission reductions as a postprocessing tool to LOTOS-EUROS)

- 5 AERONET wavelength bands

- Wave length bands that are used by POLDER

The user can select in the lotos-euros-output.rc file which parameters and wave lengths are needed to reduce unnecessary output.Internally all parameters are calculated for all wavelengths in the set. When no optical properties are needed, the switch *with_radiation* in the main rc-file can be set to *False*. This is the default value.

For every population (fine dust and nondust, coarse dust and nondust), the dimensionless extinction efficiency ext is extracted from the look-up table for the fine or coarse mode. From that, the extinction cross section normalized to a single particle is calculated by multiplying it by the square of the wave length λ. The extinction coefficientis then obtained by multipling with the particle number concentration.

$$k_{ext,i} = ext_{lut}\lambda^2 N_i \tag{2.18}$$

The net **extinction coefficient** k_{net} is the sum over all modes, the optical depth per layer τ combines the extinction and the layer height, and the AOD is the sum over all modes over the length of the column

$$k_{net} = \sum_{i=1}^{4} k_{ext,i} \tag{2.19}$$

$$\tau_l = \sum_{i=1}^{4} k_{ext,i}\Delta h_l \tag{2.20}$$

$$AOD = \sum_{l=1}^{nlayers} \sum_{i=1}^{4} k_{ext,i}\Delta h_l \tag{2.21}$$

The **single scattering albedo** cannot be added linearly like AOD.It has to be calculated for the total population. Single scattering albedos ssa_i are extracted from the lookup table to obtain the scattering coefficient for the dust and nondust fine and coarse mode by

$$k_{sca,i} = k_{ext,i} * ssa_i \tag{2.22}$$

and the net single scattering albedo is

$$SSA = \frac{\sum_i k_{sca,i}}{\sum_i k_{ext,i}} \tag{2.23}$$

The **asymmetry parameter** g is also a weighed combination of the g_i values from the lookup table:

$$g = \frac{\sum k_{sca,i} g_i}{\sum k_{sca,i}} \tag{2.24}$$

2.6.4   Further steps and limitations

A few remarks should be made.Although the procedure was made with great care, it comes with limitations.

It is straightforward to estimate the (horizontal) visibility from the extinction coefficient by using it in the general relation (e.g. Seinfeld and Pandis 2006)

$$x_v = \ln(0.02)/k_{net} \tag{2.25}$$

It should be noted that visibility is also affected by scattering by gases and the presence of fog or rain, and by turbulence, which are not taken into account here.

In the current approach, only two size modes (accumulation and coarse mode) are taken into account. This approach is consistent with the AERONET division in fine and coarse mode aerosol. Depending on the application and wave length, it could be useful to define additional modes.

The optical toolbox that is currenlty used does not take into account non-sphericity of particles, whereas dust particles are in fact non-spherical. Also one has the freedom to define wave bands. For large wave length (infrared, significantly larger than 1 μm) Mie scattering by small aerosols may become less relevant.

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