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CONVERSION OF THE AEROSOL OPTICAL PROPERTIES FROM DRY TO AMBIENT RH AT THE JRC-ISPRA STATION FOR ATMOSPHERIC RESEARCH

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Outline: the study envisages the aerosol hygroscopicity, described in terms of enhancement factors for scattering, absorption, extinction and backscattering
Goal: correction of optical properties to ambient conditions → evaluate enhancement factors = $f_\chi(RH)$
Why: standardized in-situ measurements are taken in "dry" conditions (RH<30%)
Main result: extinction enhancement factor becomes larger than 2 for RH>90%, with uncertainty increasing with RH (from ~15% to ~25%).

Procedure:

-Measurements:
- -Site: EMEP-GAW station at Ispra (IT04-IPR), Italy.
- -Period analyzed: 1062 hourly data during 2008-2009
- -In-situ instruments: DMPS, APS, nephelometer, aethalometer, HTDMA
- -Input data errors: ±10% *NSD*, ± 3 % diameter (DMPS, APS), ± 1.5% aerosol scattering and backscattering (nephelometer), ± 4% aerosol absorption (aethalometer), ± 3% growth factor at 90% *RH* (HTDMA)
- -HTDMA retrievals are performed using TDMAfit software (Gysel et al. 2009).

-Mie theory

Enhancement factors:
$$f_\chi(RH,\lambda)=\frac{\chi(RH,\lambda)}{\chi(RH=0,\lambda)} \qquad (1)$$

where χ can be σ, α, κ or β, denoting the scattering, absorption, extinction or backscatter coefficient respectively. *RH* corresponds to the ambient conditions.

Methodology is sketched in the flow cart. The main steps are:
-Determine refractive index at instruments *RH,* by matching σ and α measurements and Mie calculations (Figs. 2-3).
-Determine the refractive index in dry conditions (*RH*=0%) and at ambient *RH* (Fig. 3).
-Calculate σ, α, κ or β in dry and ambient conditions
-Calculate enhancement factors (as ambient / dry) (Fig. 5)
Criteria:
1) eliminate points for which the relative error between measurements and Mie is > 30%
2) Eliminate points for which retrieved refractive index at instruments *RH* is 1.3 or 1.7 (limits on lookup table)
3) Remove outliers in the Mie-measurements regression
⇒ From 1062 points ⇒ 564 points

Error computation
- sensitivity study: there is one run using the overestimated input parameters (ε_x=+n% error) and one run using underestimated input parameters (ε_x=-n% error).

For each variable y computed along the flow chart, its uncertainty will be given by the average between the relative errors with respect to the case of ε_x=0:
$$\varepsilon_y=100\frac{1}{2}\left(\left|\frac{y_m}{y}-1\right|+\left|\frac{y_p}{y}-1\right|\right)(\%) \qquad (2)$$

y corresponds to the input parameters x (ε_x=0, i.e. no error in input parameters), while y_m and y_p correspond to the input parameters $x-\varepsilon_x$ and $x+\varepsilon_x$ respectively

Results

- at high *RH* (>90%) enhancement factors can reach values of ~ 6, 5,1.2 and 4 for σ, α, κ and β (Fig. 5); a seasonal/diurnal behaviour is expected
- very good correlation between enhancement (except α) factors and growth factors (Fig. 6)
- large difference for *g* (asymmetry param.) between empirical formula and Mie calculations (Fig. 4)
- errors (e.g. 10.02.2008)
  - -There is a strong correlation between the error and *RH* for the imaginary part of refractive index, GF, σ, κ, β, f_χ(RH) for χ = σ, κ, and β (Fig. 8)
  - -Average error for other variables are shown in Table 1. Note also that the largest error occur for the imaginary part of refractive index

Conclusions

The scheme involved to determine enhancement factors can be used to correct for the optical measurements taken in dry conditions: knowing the aerosol *GF(RH)*, one applies the relationship between *f*(*RH*) and *GF*(*RH*) (Fig. 6) and obtain *f*(*RH*). Then, apply eq. (1) to get σ, α, κ or β at *RH*. A careful estimation of the uncertainty is necessary. The estimation of the weights of each particular input error is under investigation.

References

Andrews, E., et al.: Comparison of methods for deriving aerosol asymmetry parameter, *J. Geophys. Res.*, 111, D05S04, doi:10.1029/2004JD005734, 2006.
Gysel, M., et al., Inversion of tandem differential mobility analyser (TDMA) measurements, *J. Aerosol Sci.*, 40, 134-151, 2009.

Fig. 2. σ, κ, α, β Mie vs measurements

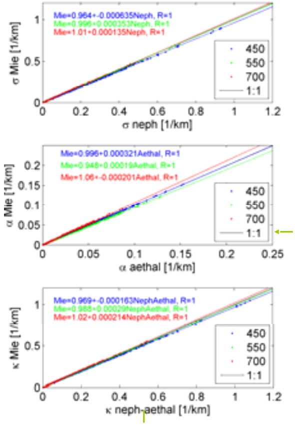
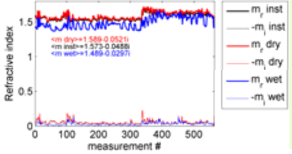


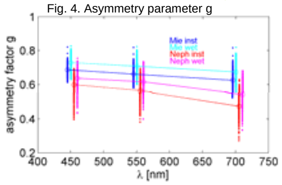
Fig. 3. Refractive index



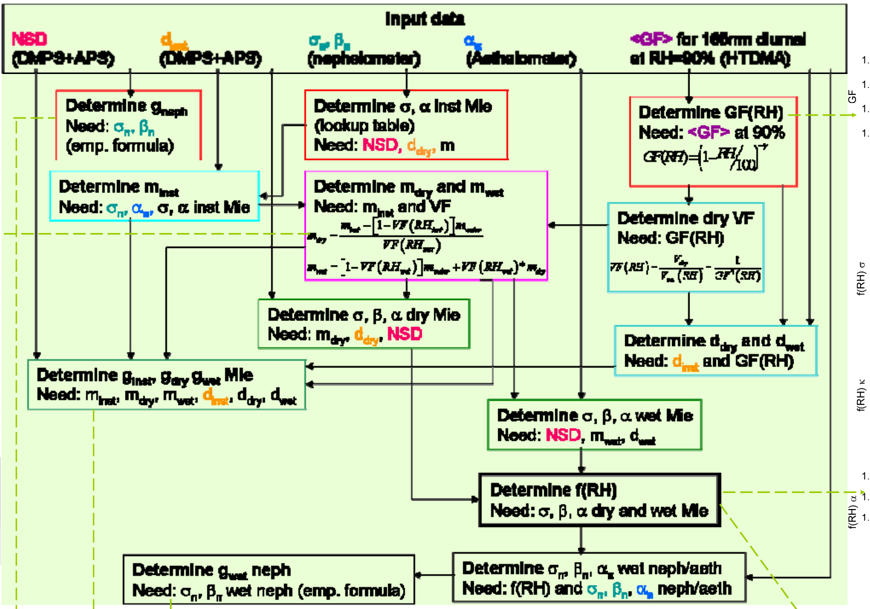
<g>	450	550	700 nm
Neph inst	0.60	0.57	0.48
Mie inst	0.69	0.66	0.63
Neph wet	0.64	0.62	0.55
Mie wet	0.73	0.71	0.68

Rel diff w.r.t. neph (%)			
Inst:	14.60	16.73	30.89
Wet:	14.42	14.70	23.52

Fig. 4. Asymmetry parameter g



Flow chart to determine enhancement factors *f*(*RH*)



Input data
NSD (DMPS+APS)
d_{inst} (DMPS+APS)
σ_n, β_n (nephelometer)
α_a (Aethalometer)
<GF> for 165nm diurnal at RH=90% (HTDMA)

Determine g_{neph}
Need: σ_n, β_n (emp. formula)

Determine σ, α inst Mie (lookup table)
Need: NSD, d_{dry}, m

Determine GF(RH)
Need: <GF> at 90%
$GF(RH)=\left(1-RH/100\right)^{-\gamma}$

Determine m_{inst}
Need: σ_n, α_a, σ, α inst Mie

Determine m_{dry} and m_{wet}
Need: m_{inst} and VF
$m_{dry}=\frac{m_{inst}-[1-VF(RH_{inst})]m_{water}}{VF(RH_{inst})}$
$m_{wet}=[1-VF(RH_{wet})]m_{water}+VF(RH_{wet})*m_{dry}$

Determine dry VF
Need: GF(RH)
$VF(RH)=\frac{V_{dry}}{V_{wet}(RH)}=\frac{1}{GF^3(RH)}$

Determine σ, β, α dry Mie
Need: m_{dry}, d_{dry}, NSD

Determine d_{dry} and d_{wet}
Need: d_{inst} and GF(RH)

Determine g_{inst}, g_{dry} g_{wet} Mie
Need: m_{inst}, m_{dry}, m_{wet}, d_{inst}, d_{dry}, d_{wet}

Determine σ, β, α wet Mie
Need: NSD, m_{wet}, d_{wet}

Determine f(RH)
Need: σ, β, α dry and wet Mie

Determine g_{wet} neph
Need: σ_n, β_n wet neph (emp. formula)

Determine σ_n, β_n, α_a wet neph/aeth
Need: f(RH) and σ_n, β_n, α_a neph/aeth

NSD = particles number size distribution (DMPS+APS)
d_{inst}, d_{dry}, d_{wet} = particles diameter at instruments *RH*, *RH*=0% and ambient *RH* (DMPS+APS)
σ_n = aerosol scattering coefficient (nephelometer)
β_n = aerosol backscatter coefficient (nephelometer)
α_a = aerosol absorption coefficient (aethalometer)
<*GF*> 165nm diurnal = average *GF* (monthly diurnal) for 165nm at *RH*=90%
G_{neph} = asymmetry parameter empirically determined from nephelometer
g_{inst}, g_{dry}, g_{wet} Mie = asymmetry parameter at instruments *RH, RH*=0% and ambient *RH* computed with Mie theory
m_{inst}, m_{dry}, m_{wet} = refractive index at instruments *RH*, *RH*=0% and ambient *RH*
VF=dry volume fraction
f(*RH*)=enhancement factor for each of σ, κ, α, β
emp. formula = by Arnott (Andrews et. Al, 2006)
Note: σ_n, β_n, α_a neph/aeth are not measured at RH=0%.

Errors. Example for 10.02.2009.

Table 1. The mean errors (%) for variables not showing a RH dependence.

	$m_{r,dry}$	$m_{r,inst}$	$m_{r,wet}$	$m_{i,dry}$	$m_{i,inst}$	$g_{Mie,inst}$	$g_{Mie,dry}$	$g_{Mie,wet}$	GF_{inst}	GF_{dry}	$\sigma_{Mie,dry}$	$\alpha_{Mie,dry}$	$\kappa_{Mie,dry}$	$\beta_{Mie,dry}$
Error [%]	4.84	4.65	3.03	11.20	11.92	3.47	3.55	2.83	0.24	2.63	1.34	4.44	1.46	2.85

The largest errors occur for the imaginary part of the refractive index (Fig. 7).
This is driven by the error in *VF*, which is ~ error in GF^3.
⇒large errors in σ, κ, β ⇒ large errors in $f_\chi(RH)$ for
χ = σ, κ, β (Fig. 8)

Fig. 7. Errors for refractive index

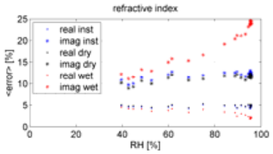


Fig.8. Errors for refractive index, *GF* and *f*(*RH*)

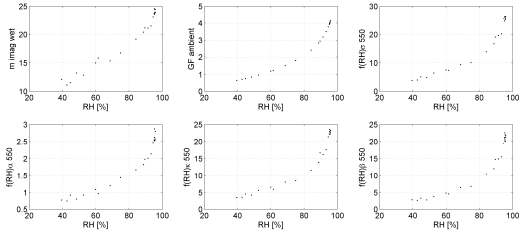


Fig. 1. *GF*(*RH*)

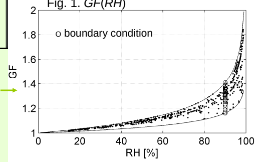


Fig. 5. *f*(*RH*) λ=550nm

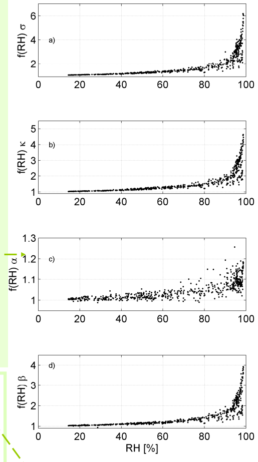
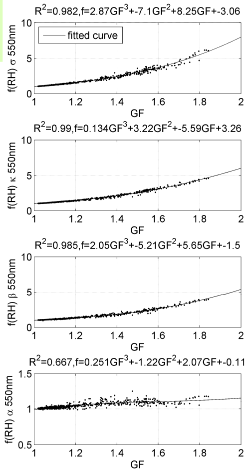


Fig. 6. *f*(*RH*) vs *GF*(*RH*)


R^2=0.982,f=2.87GF^3+-7.1GF^2+8.25GF+-3.06
R^2=0.99,f=0.134GF^3+3.22GF^2+-5.59GF+3.26
R^2=0.985,f=2.05GF^3+-5.21GF^2+5.65GF+-1.5
R^2=0.667,f=0.251GF^3+-1.22GF^2+2.07GF+-0.11