KU LEUVEN

MeBiOS

Optical characterization of the composition and scatterer size distributions of turbid liquids from Vis/NIR spectroscopy

W. Saeys, A. Postelmans, R. Watté, R. Van Beers, B. Aernouts
Wouter.saeys@kuleuven.be

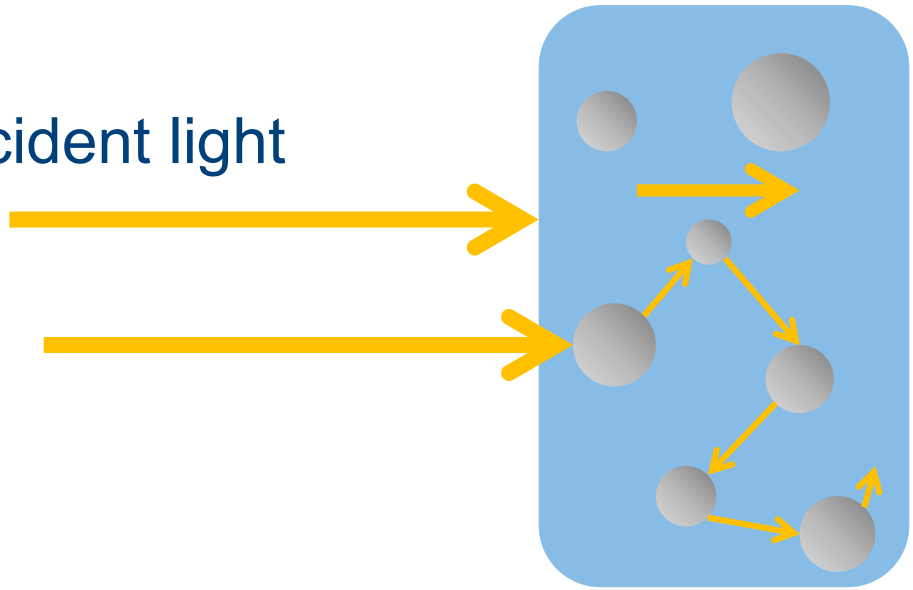
Overview

- Introduction
- From optical measurements to bulk optical properties
  - Double integrating spheres
  - Spatially resolved spectroscopy
- From scattering spectra to particle size distribution
  - Shape dependent
  - Shape independent
  - Case study polystyrene particles
- Conclusions

Introduction

Light propagation in turbid media

Incident light



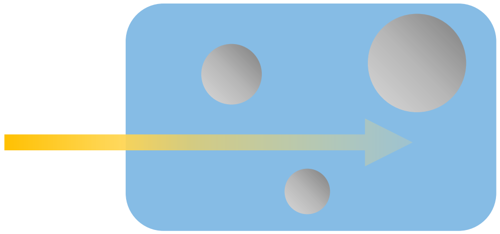
Absorption

Scattering

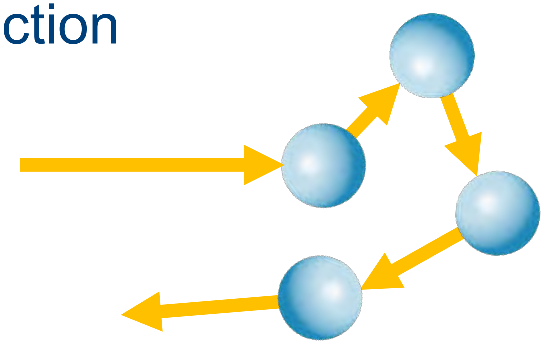
KU LEUVEN

Bulk scattering and absorption coefficient

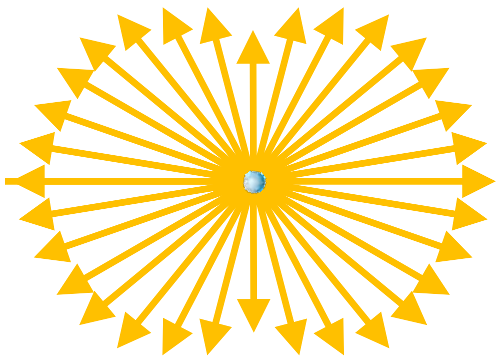
- Bulk absorption coefficient μ_a
  - Probability of photon absorption per unit infinitesimal pathlength



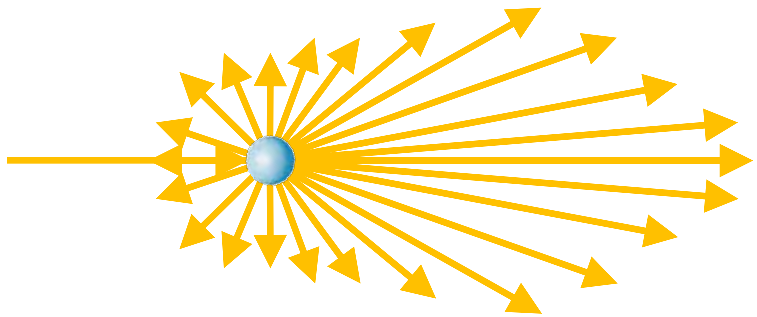
- Bulk scattering coefficient μ_s
  - Probability of photon scattering per unit infinitesimal pathlength
  - Non-linear effect on light extinction



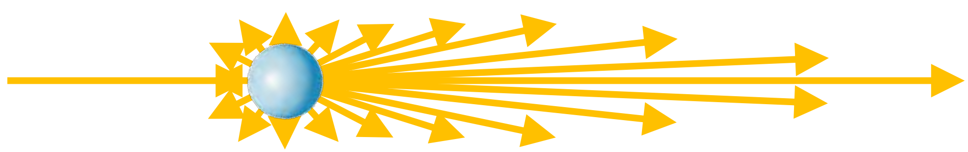
Anisotropy factor *g*



Particle << wavelength
Anisotropy = 0
Isotrope scattering



Particle ≈ wavelength
Anisotropy ≈ 0.6



Particle > wavelength
Anisotropy → 1

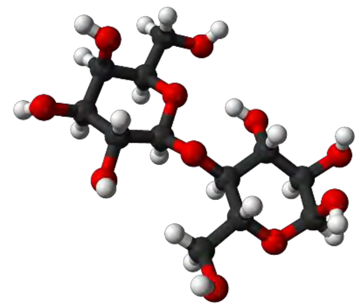
Optical properties and product characteristics

- Why interest in bulk optical properties?
  → Related to emulsion/suspension characteristics

Chemical

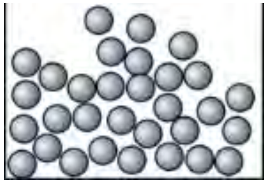
μ_a

Bulk optical properties



Chemical composition

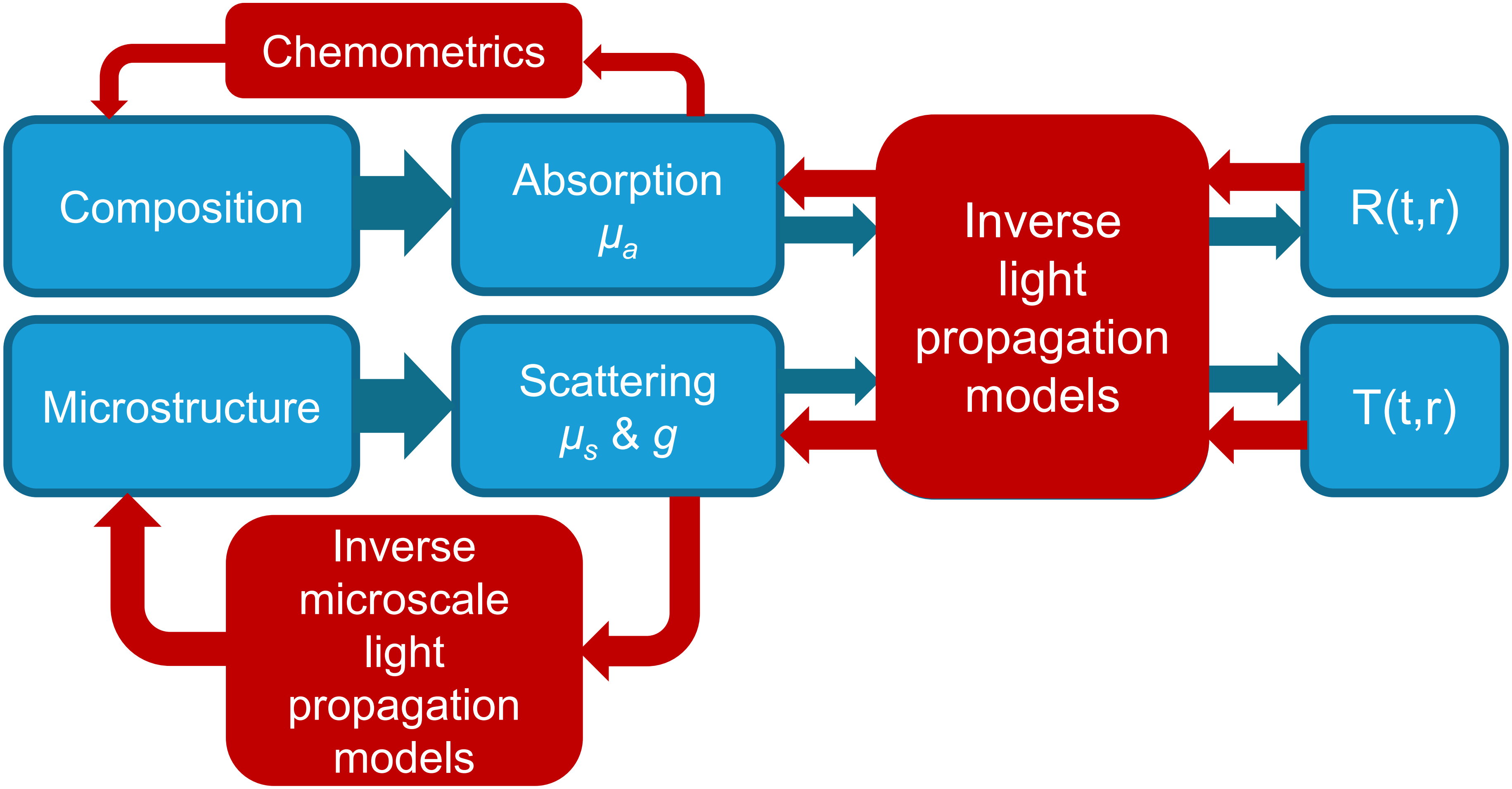
μ_s



Particle size distribution
Volume fraction

Physical

Research hypothesis



Bulk optical properties

From optical measurements to BOP

KU LEUVEN

BOP from optical measurements

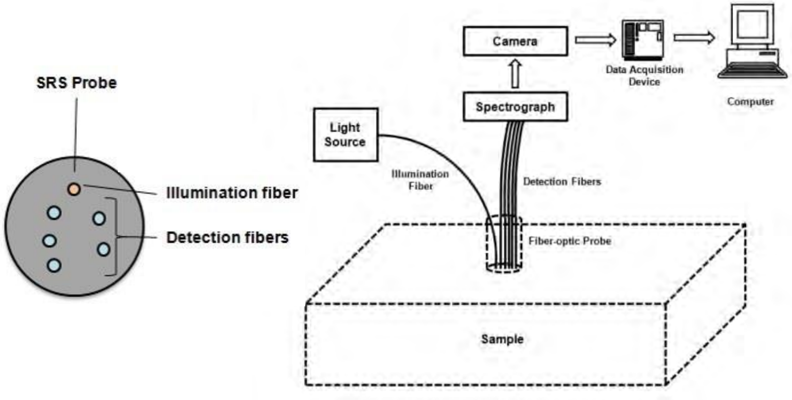
- Calculate BOP from multiple (uncorrelated) measurements

Reflectance & transmittance

- o Double integrating spheres (DIS)
- o Unscattered transmission (UT)

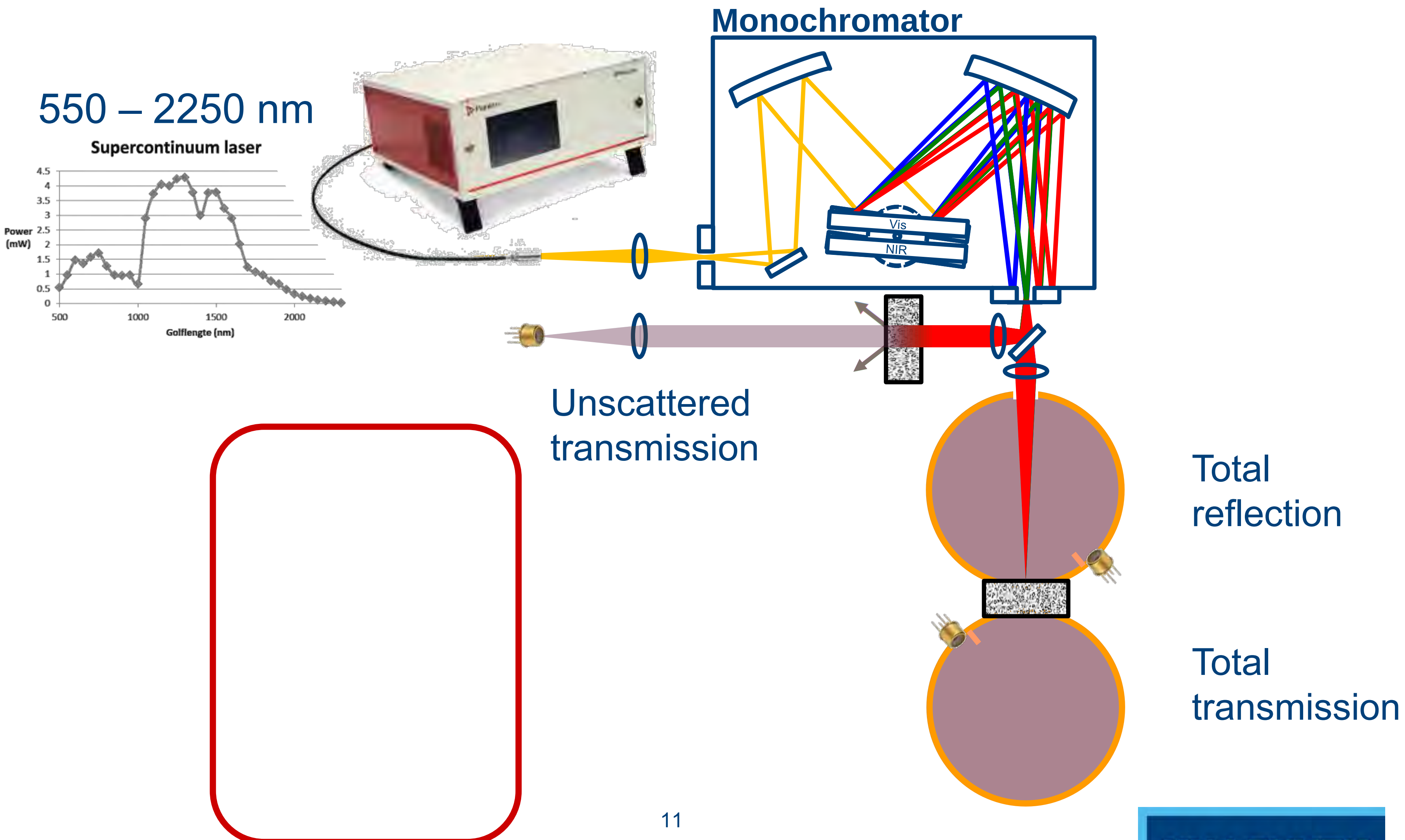
Multiple distances from light source

- o Spatially resolved spectroscopy (SRS)



KU LEUVEN

Double integrating spheres set-up



Aernouts *et al.*, Opt. Express 21 (2013)

Inverse adding-doubling (IAD)

- Find optical properties that correspond to measured reflection and transmission

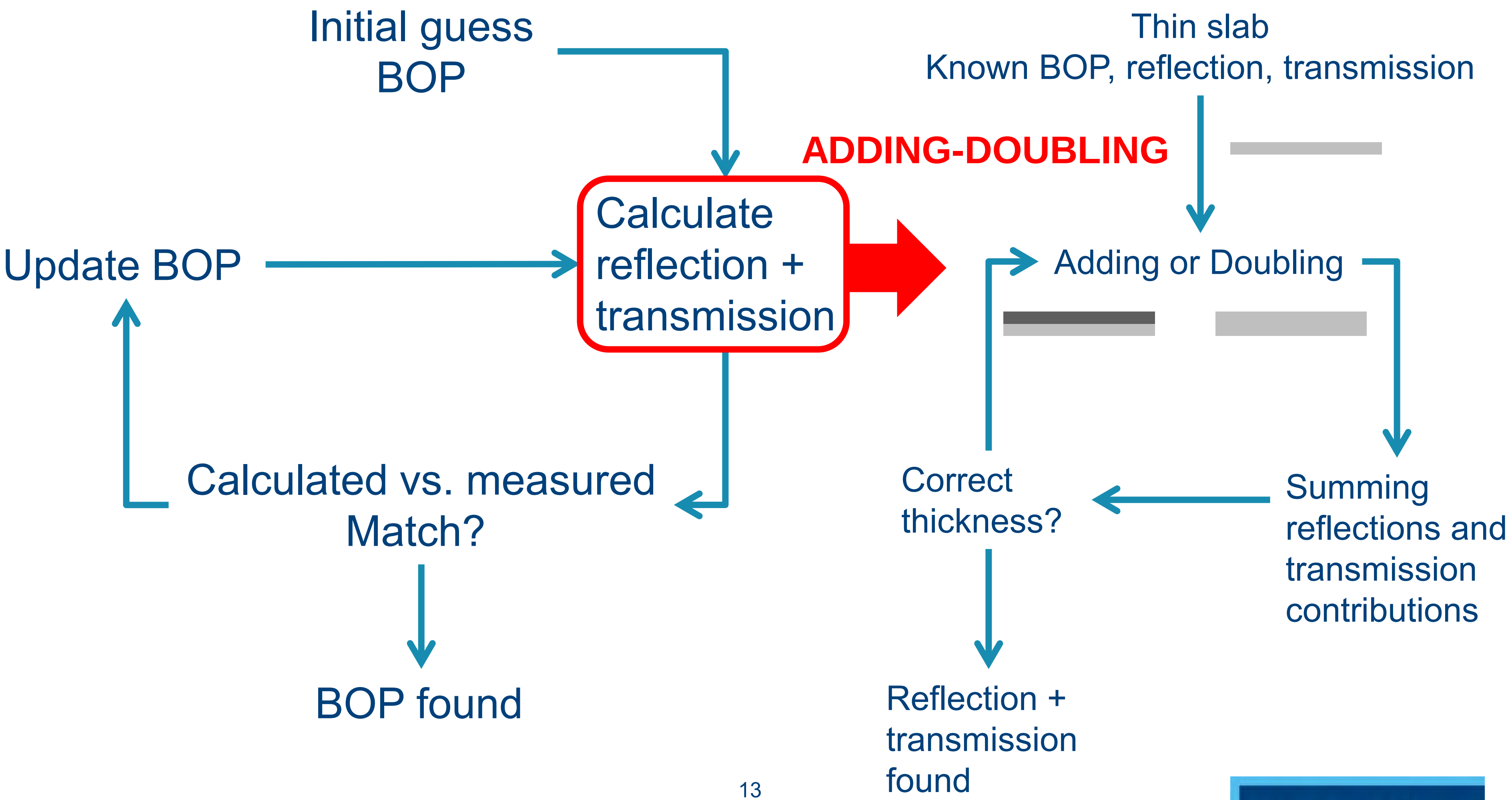
- 3 optical measurements
  - Diffuse reflection
  - Diffuse transmission
  - Unscattered transmission

Diffuse reflection + Diffuse transmission → μ_a, μ_s'

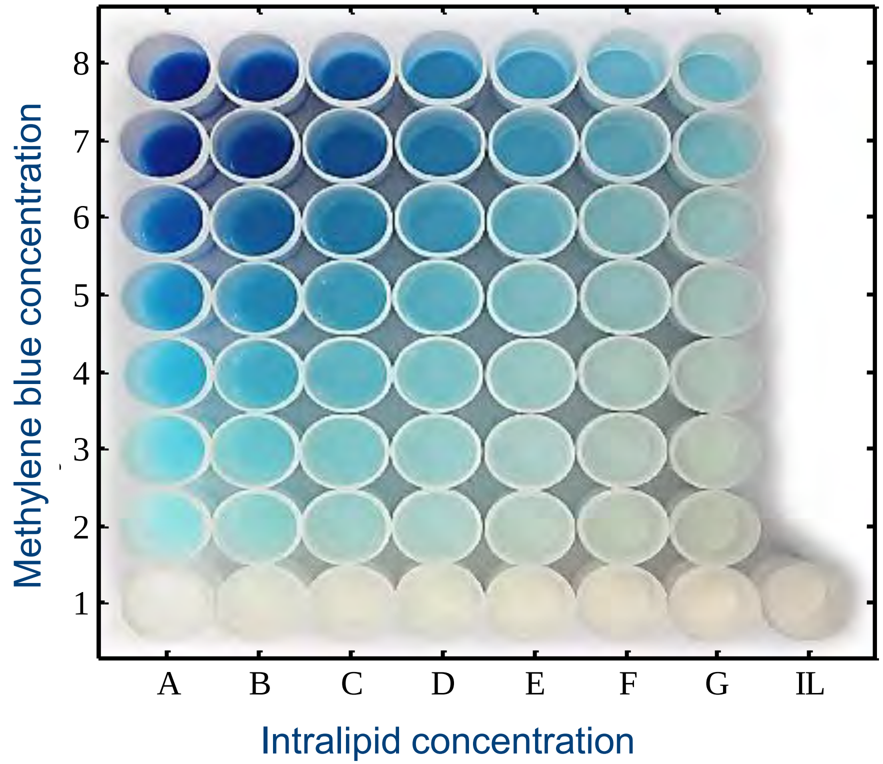
Diffuse reflection + Diffuse transmission + Unscattered transmission → μ_a, μ_s, g

- Iterative process

Inverse adding-doubling (IAD)

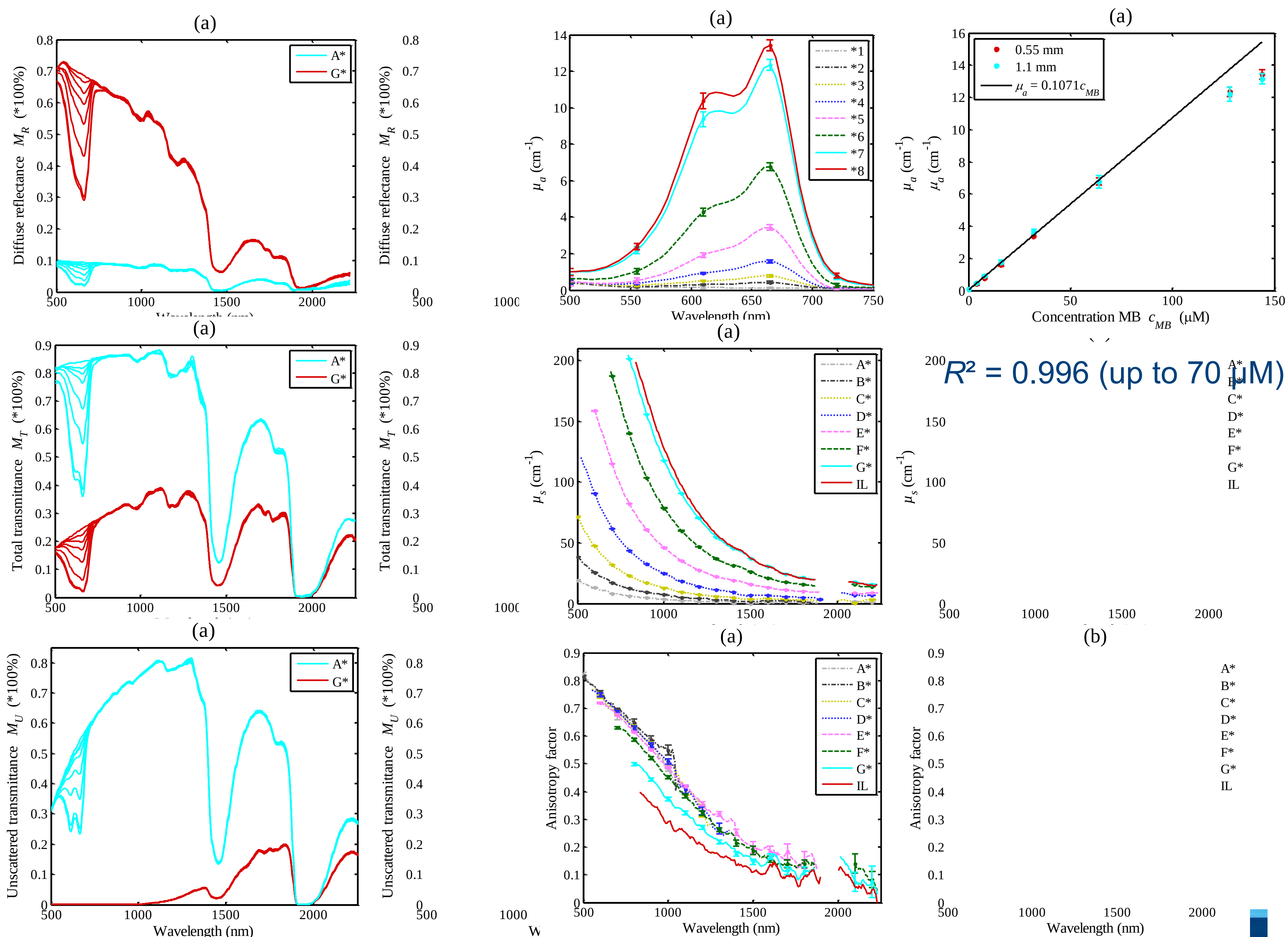


Example: optical phantoms



KU LEUVEN

Aernouts *et al.*, Opt. Express 21 (2013)

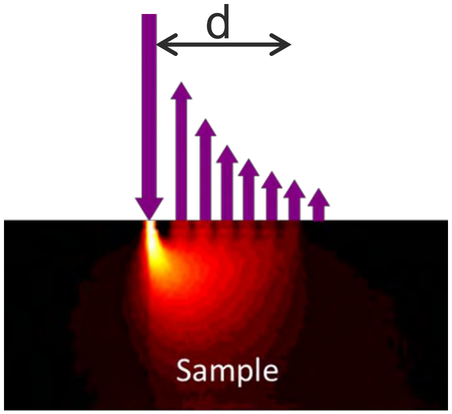


$R^2 = 0.996$ (up to 70 µM)

Aernouts *et al.*, Opt. Express 21 (2013)

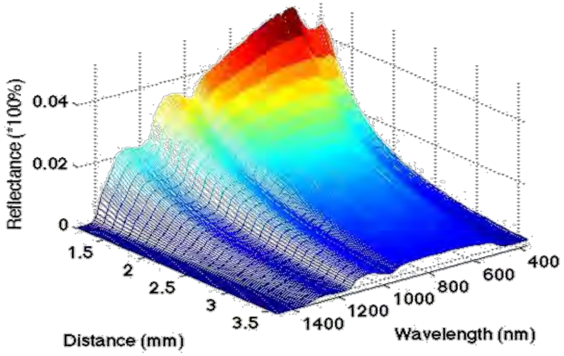
Spatially resolved spectroscopy (SRS)

- Detectors at multiple distances from light source



- Interaction history is function of distance d
  - o Further from light source
    - Lower signal
    - More interaction with tissue

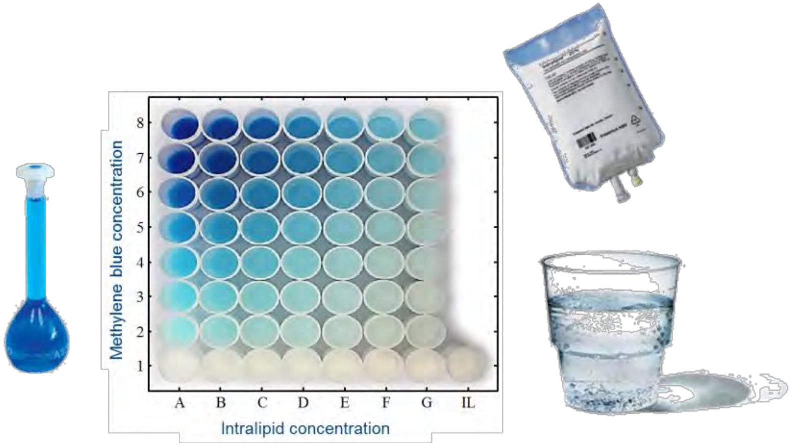
- Intensity profile R(d)



- Possible for dense samples without dilution

Estimate BOP from SRS data

- Forward light propagation model
  - Adding-doubling → no 2D information
  - Diffusion approximation → assumptions not valid
  - Monte Carlo simulations → computationally expensive
  - → Data-based metamodeling approach
    - Stochastic Kriging
    - Train on set of liquid phantoms covering wide range of BOP

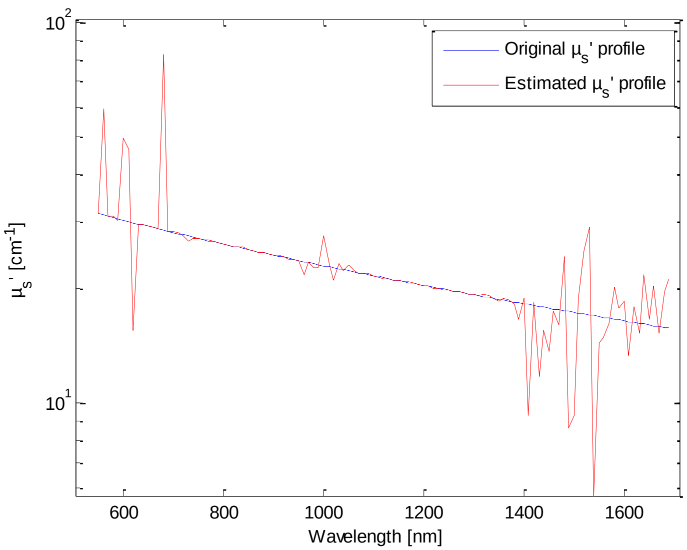


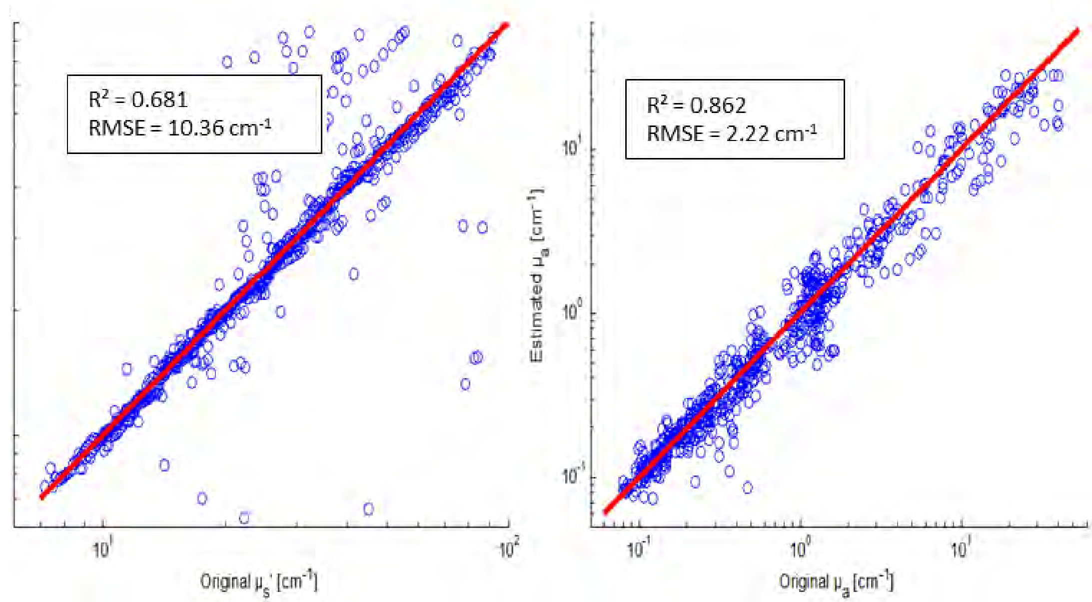
Watté *et al.*, Opt. Express 21 (2013)

KU LEUVEN

Wavelength by wavelength

- Iterative optical properties estimation
  - o Nelder-Mead optimizer for minimization
    - Cost function = sum of squared relative errors
  - o No assumptions on scattering or absorption profiles used





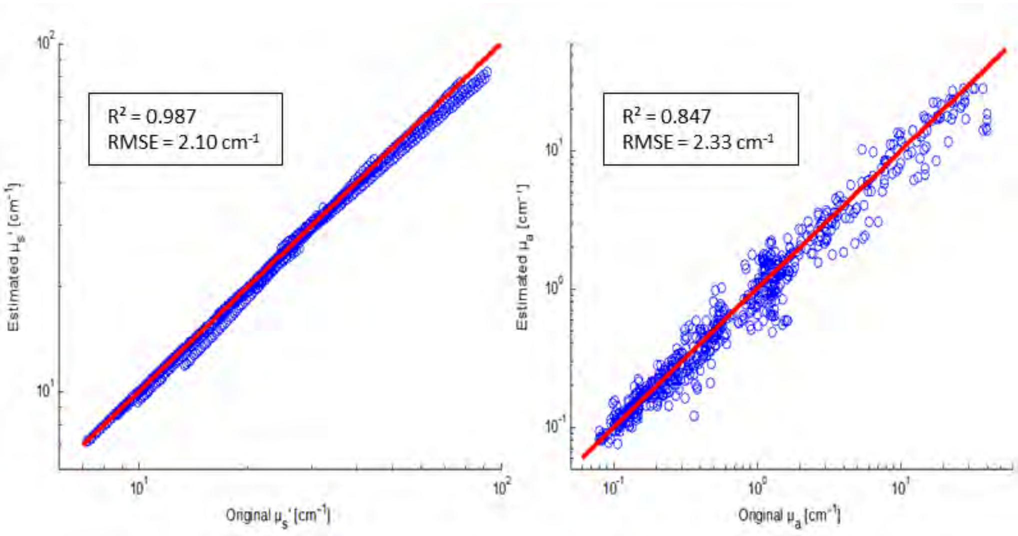
Watté *et al.*, Opt. Express 21 (2013)

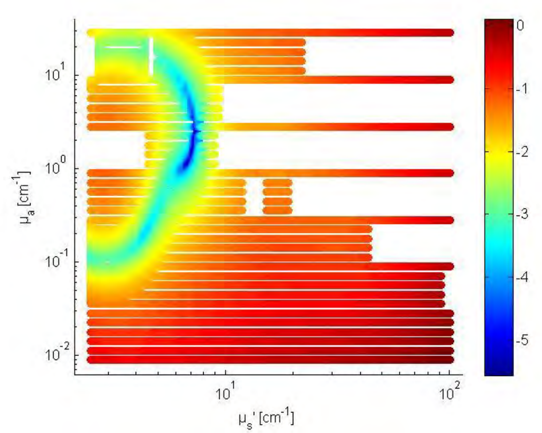
Constrained optimization

- Include expert knowledge: μ_s' as parametric function
  - Trade-off smoothness - flexibility

$$\mu_s'(WL) = p_1.exp(p_2.WL) + p_3 + p_4.WL + p_5.WL^2 + p_6.WL^3$$

- Minimising cost function over entire wavelength range
- Construction of 'information grid' to select best combination





KU LEUVEN

Watté *et al.*, Opt. Express (2016)

Particle size distribution estimation

From scattering spectra to PSD

KU LEUVEN

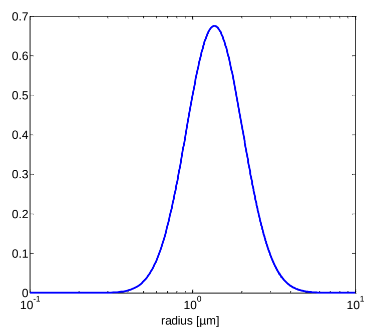
Forward problem

- Calculate optical properties for known PSD

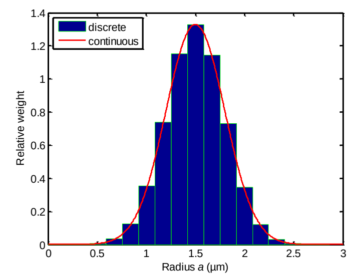
MICROSCALE

Physical information

- Particle size distribution
- Volume fraction scatterers
- Refractive indices

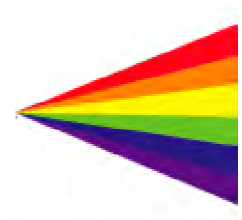


Discretise PSD



+

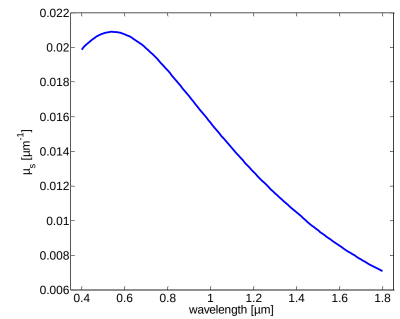
Mie theory



MACROSCALE

Bulk optical properties

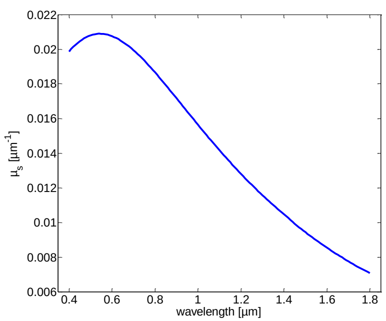
- Bulk absorption coefficient
- Bulk scattering coefficient
- Anisotropy factor



KU LEUVEN

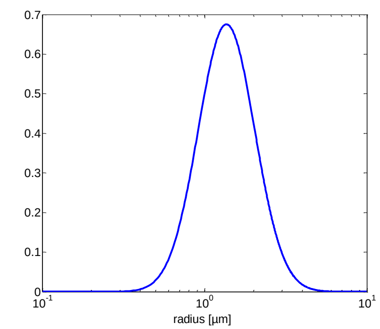
Inverse estimation PSD

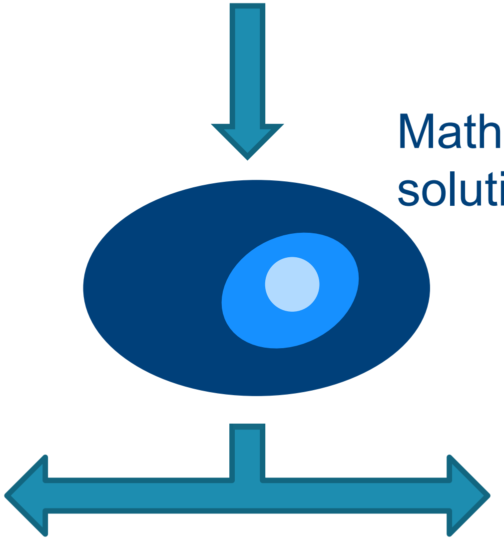
Scattering spectra
μ_s, μ_s' or g



Inverse estimation polydisperse PSD

Particle Size Distribution (PSD)
Volume fraction





Mathematical solutions

Non-negative solutions

Conform literature/ measurements

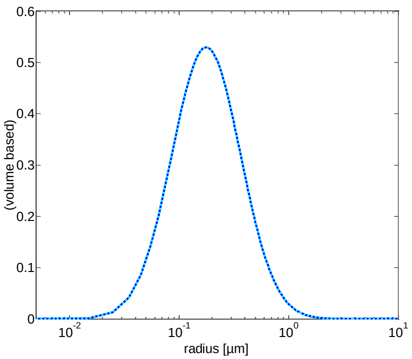
Shape dependent
→ Assume shape

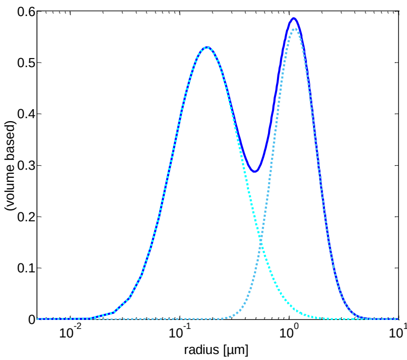
Regularization

Shape independent
→ Smoothness condition

Shape dependent PSD estimation

- Assume probability density function
  - o Monomodal: PSD = logn(μ_1,σ_1)
  - o Bimodal: PSD = scale . logn(μ_1,σ_1) + (1-scale) . logn(μ_2,σ_2)





Estimate parameter values and volume fraction

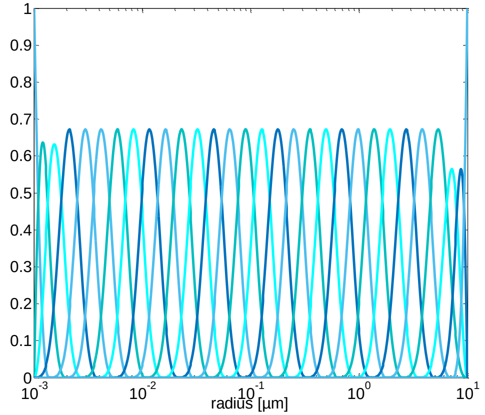
Minimize sum of relative least squared errors

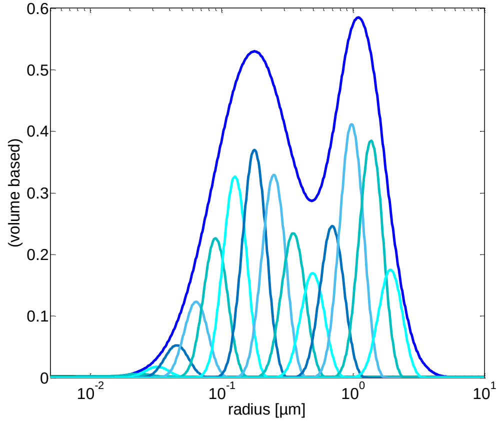
$$\min \sum_{i=1}^{N_\lambda} \left(\frac{\mu_{s,i} - \widehat{\mu_{s,i}}}{\mu_{s,i}} \right)^2$$

- Robust against noise
- Limited flexibility

Shape independent PSD estimation

- Approximate PSD by weighted sum of splines





B-spline basis → Particle radius range; Number of splines

Weighted sum B-splines

- Find B-spline weights

$$\mu_s(\lambda) = \int_{r_{min}}^{r_{max}} PSD(r).\sigma_s(r,\lambda)dr = \int_{r_{min}}^{r_{max}} \sum_{j=1}^{NB} w_j.B_j(r).\sigma_s(r,\lambda)dr = \sum_{j=1}^{NB} w_j.\mu_{s,j}(\lambda)$$

  - Calculate volume fraction from weights

Shape independent PSD estimation

- Tikhonov regularization
  - Non-negative least squares

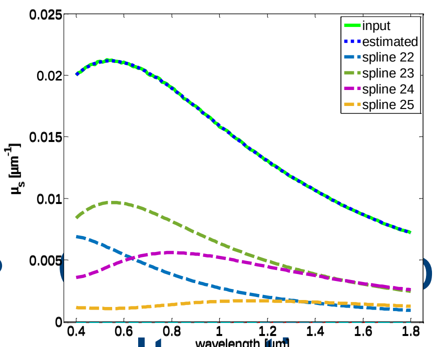
$$\min \|Aw - \mu_s\|^2 + \gamma \|Lw\|^2, \qquad w \geq 0$$

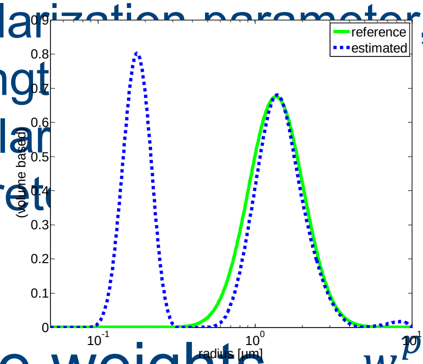
$\mu_s = \sum w_i \cdot \mu_{s,\,\text{spline } i}$

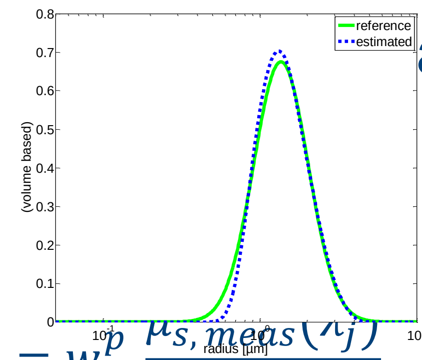
Non-smoothness penalty

γ: regulari… parameter,
strengt…
L: regular…
discret…

- More flexible
- Less robust,
…ation







- …orithm
  - Iterative update spline weights $w_j^{p+1} = w_j^p \cdot \frac{\mu_{s,meas}(\lambda_j)}{\mu^p_{s,calc}(\lambda_j)}$

KU LEUVEN

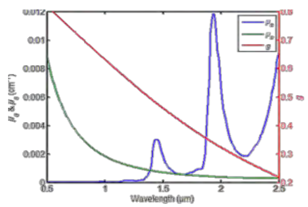
Case study polystyrene particles

Check

Reference PSD + Measured BOP

Laser diffraction
DLS

DIS + UT
500-1850 nm



IAD → BOP

Measured data

Input scattering spectrum → Estimated PSD

Inverse estimation

Simulated data

Estimated BOP

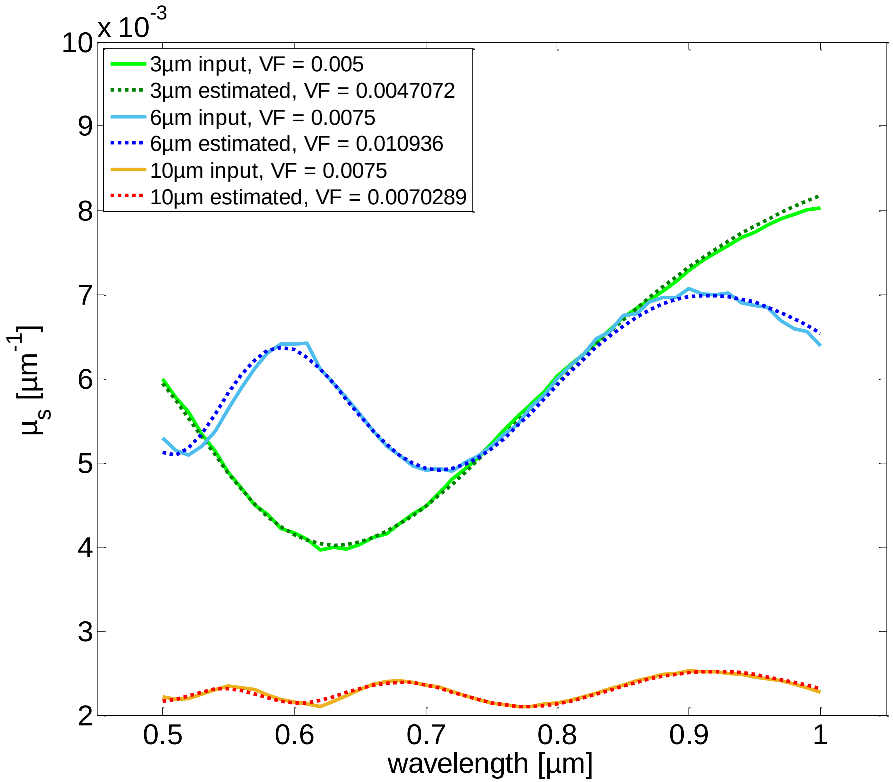
Input PSD → BOP

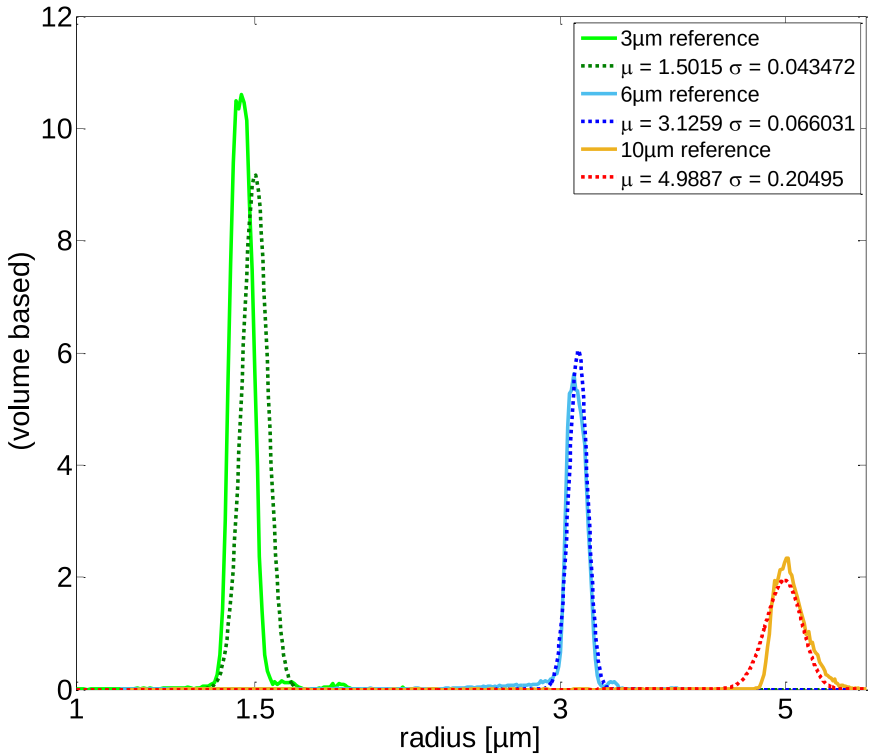
Forward simulation

Add noise

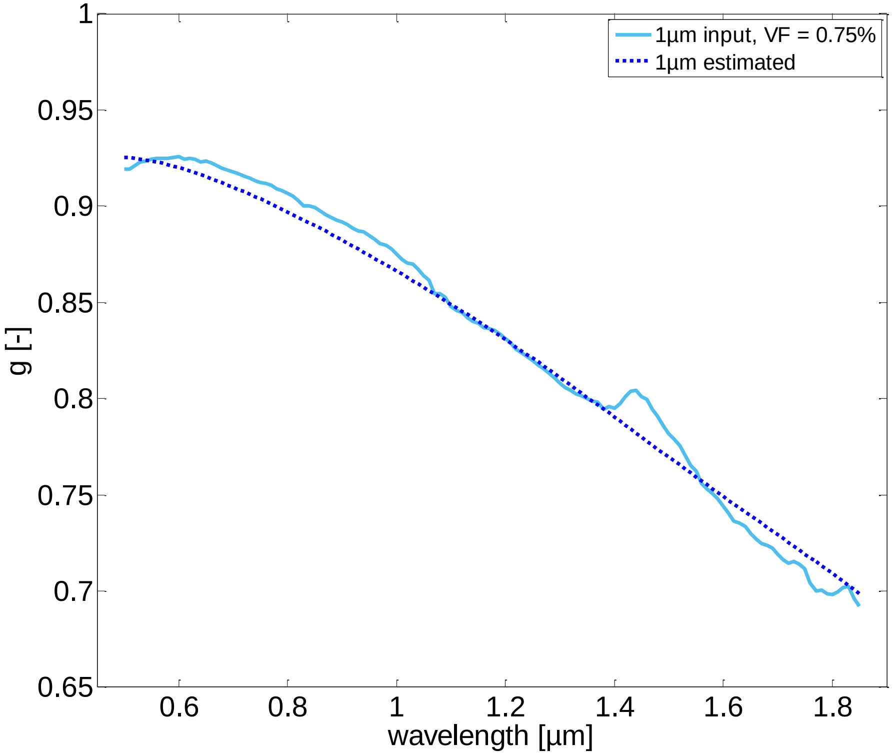
Check

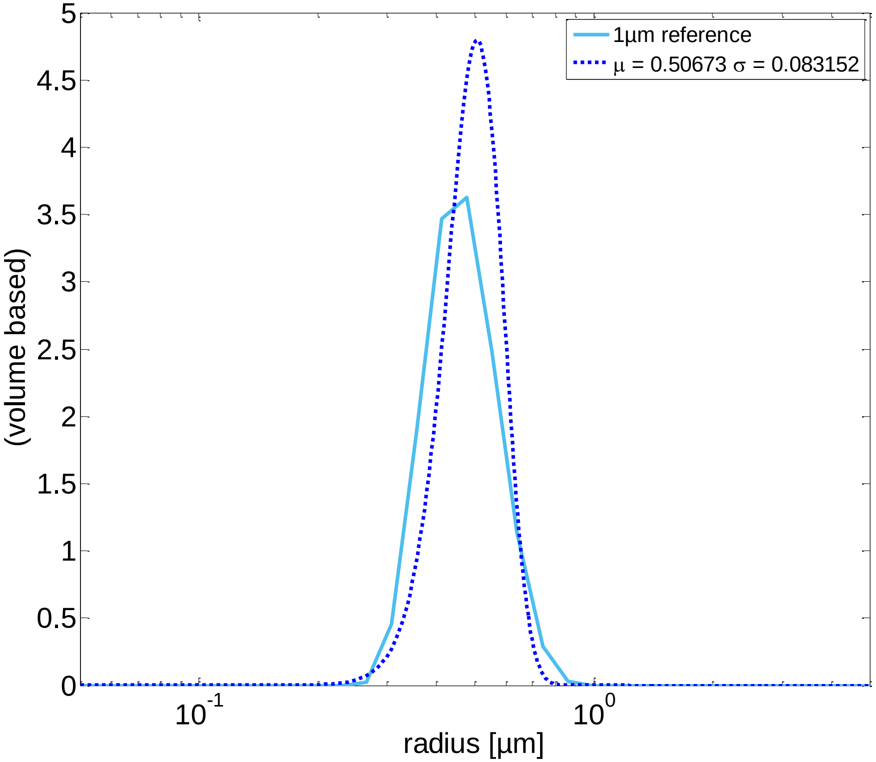
Results polystyrene
Monomodal shape dependent





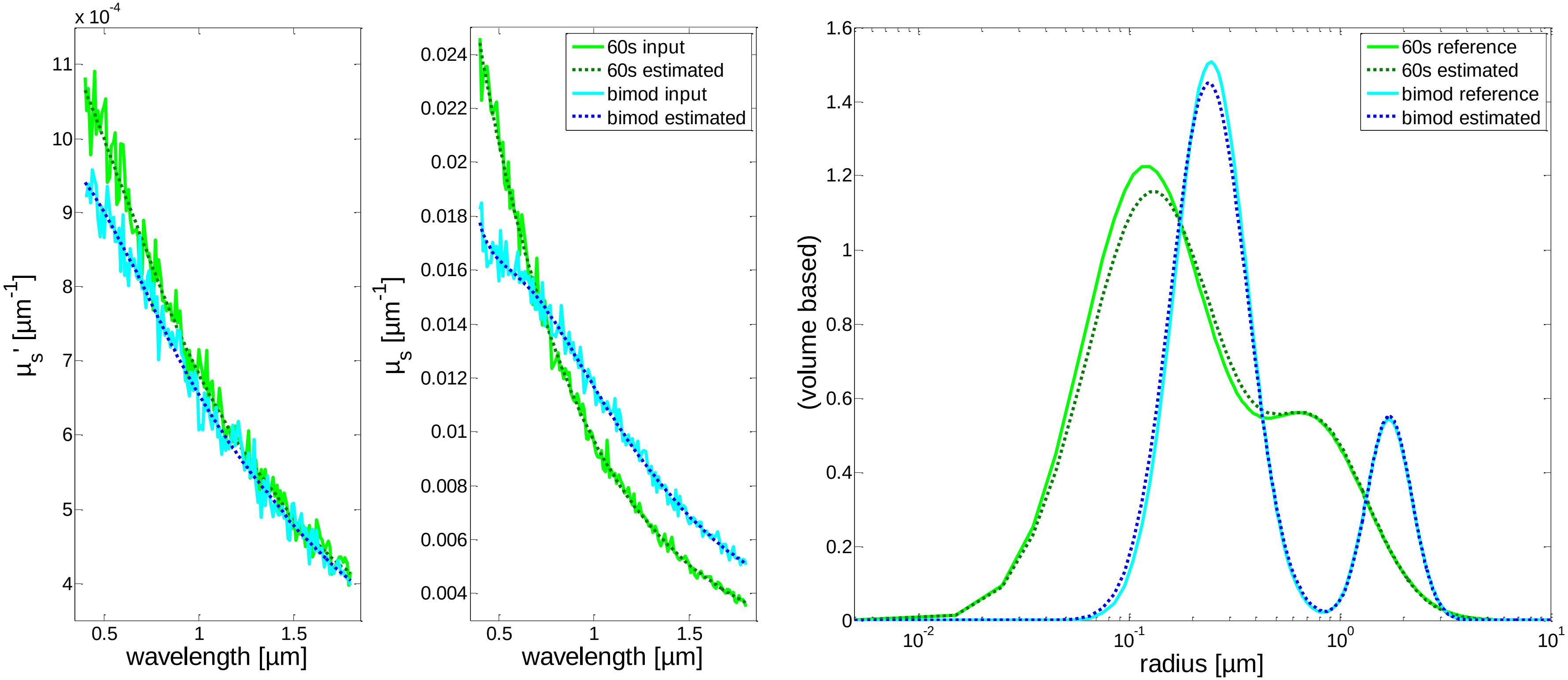
Results polystyrene
Monomodal shape dependent





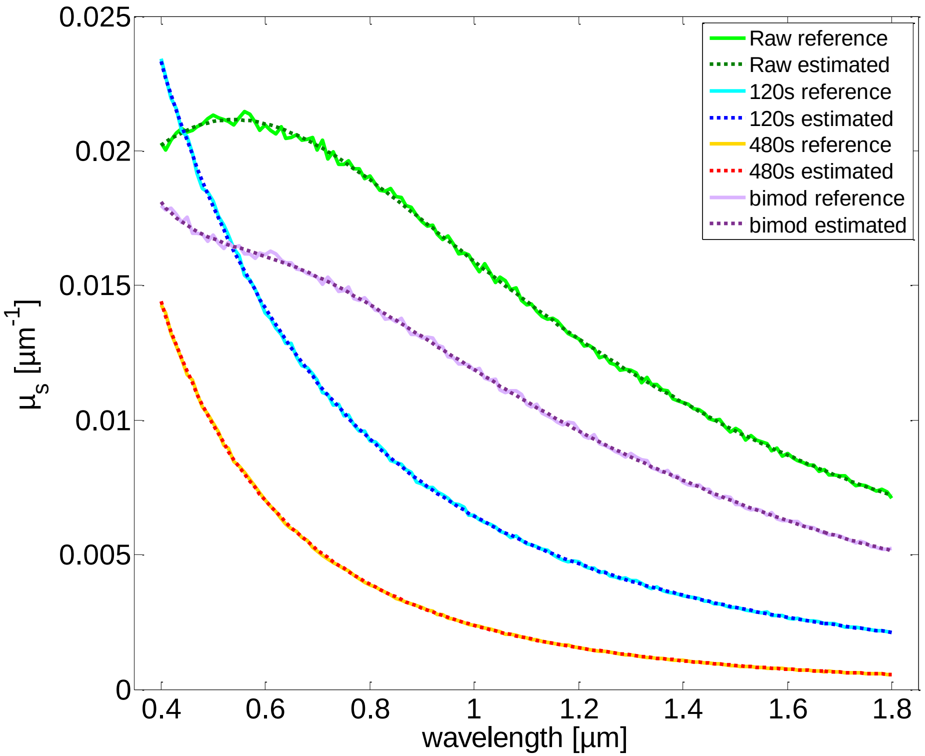
KU LEUVEN

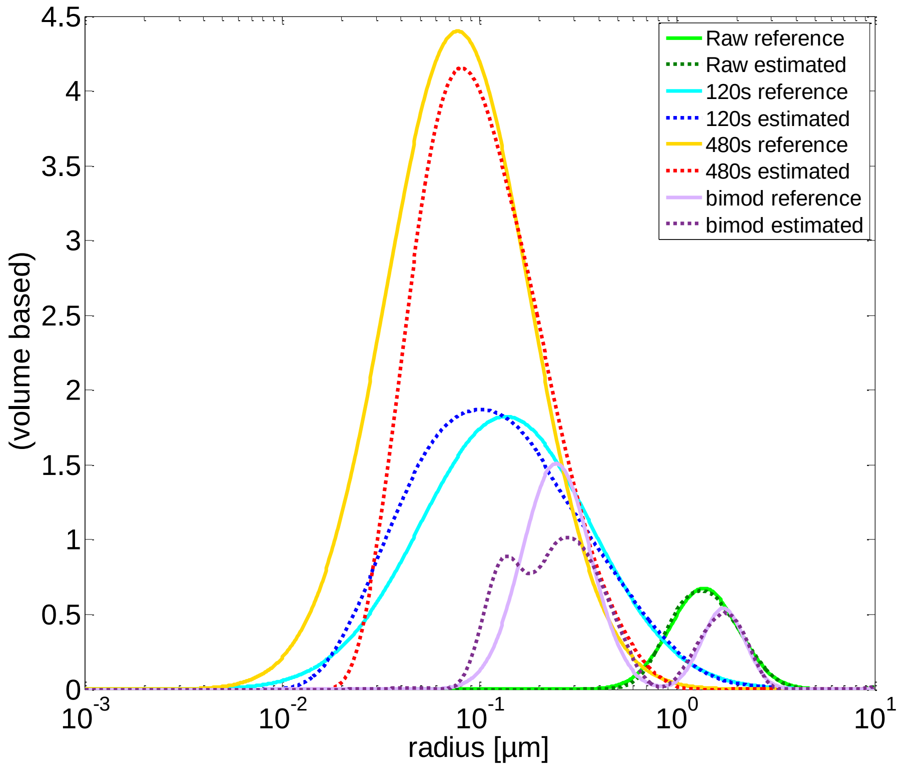
Results simulated fat in water
Bimodal shape dependent bimodal



KU LEUVEN

Results simulated fat in water
Shape independent





KU LEUVEN

Applications

- Link to product properties and quality attributes
  - Viscosity, creaming, mouth feeling/creaminess perception, nutrient uptake...
- Quality monitoring during production and storage

KU LEUVEN

Conclusions

- Accurate determination of bulk optical properties from
  - Reflectance & transmittance data
    - DIS + UT
  - Multiple reflectance measurements
    - SRS

- Use of BOP for characterizing emulsions/suspensions
  - Absorption: chemical composition
  - Scattering: PSD, volume fraction scatterers

Conclusions

- PSD estimation from Vis/NIR bulk scattering spectra
- Shape dependent method
  - Good estimation if correct choice of probability density function
- Shape independent method
  - Flexible, but more prone to artefacts
  - Good estimation if good regularization and choice B-spline basis
- Opportunities for (on-line) optical determination of microphysical emulsion/suspension quality

Questions?

Contact:
www.biophotonics.be
Wouter.saeys@kuleuven.be

KU LEUVEN