The Effect of Aerosol Swelling on the Fraction of Accumulation Mode Aerosols and the Angstrom Exponent

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Special thanks: Norman Loeb and Jim Coakley

Motivation

Loeb Study: MODIS retrievals of aerosol Angstrom exponent and fine mode fraction are greater in regions near clouds.

$$\text{Angstrom Exp: } < \alpha > \sim 0.52,\ \delta\alpha \sim 0.2$$

$$\text{Fine Mode Fraction: } < \eta > \sim 0.4,\ \ \delta\eta \sim 0.06$$

$$\text{Cloud Fraction: } < f > \sim 0.52,\ \delta f \sim 0.14$$

Does aerosol swelling play a role?

Review

Angstrom exponent (α): $\tau = \beta\lambda^{-\alpha}$

$\alpha \lesssim 1 \Rightarrow$ Size distributions dominated by the coarse mode $(r \gtrsim 0.5\,\mu m)$

$\alpha \gtrsim 2 \Rightarrow$ Size distributions dominated by the fine mode

JGR: Eck (1999), Kaufman (1994), Westphal (1991), Schuster (2006)

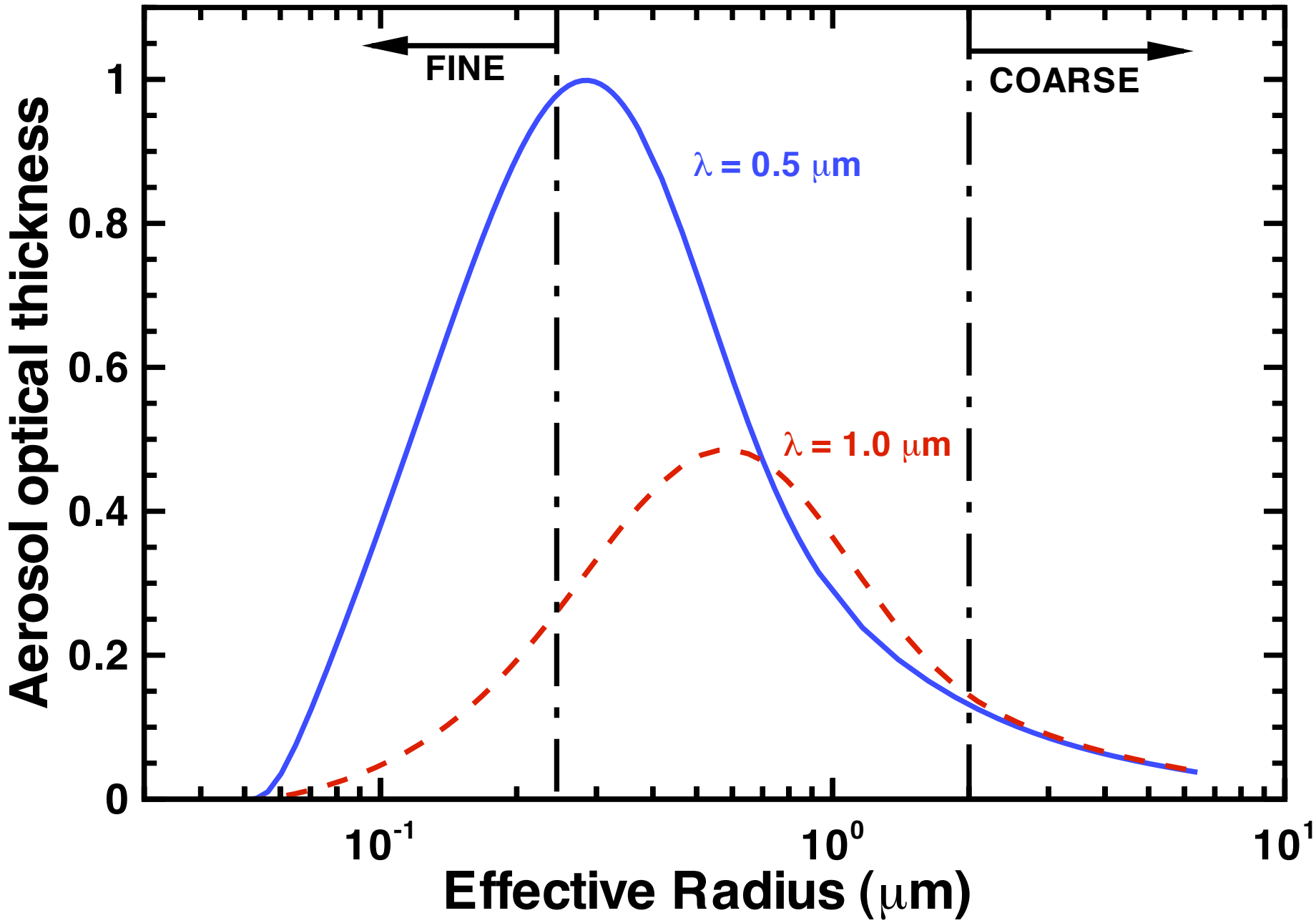
MODIS fine mode fraction:

$$FMF = \frac{AOD_{550}(fine)}{AOD_{550}}$$

Levy, JGR (2003); Remer, JAS (2005)

$$\tau = \beta\lambda^{-\alpha}$$

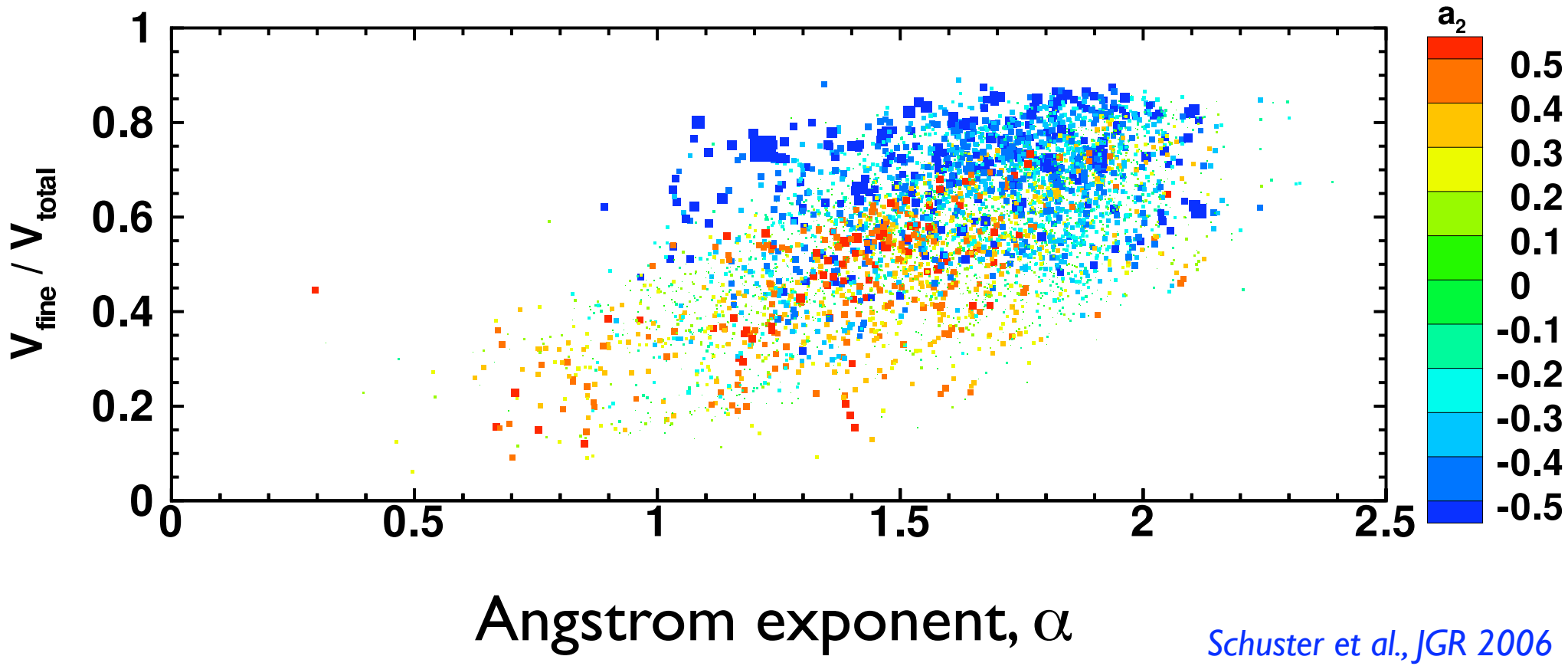
$$\ln \sigma_g = 0.38$$



Schuster et al., JGR 2006

Sensitivity of Angstrom exponent to the volume fraction of aerosols with radii less than 0.6 μm

AERONET size distributions at 53 sites in 2000 and 2001



Schuster et al., JGR 2006

Review

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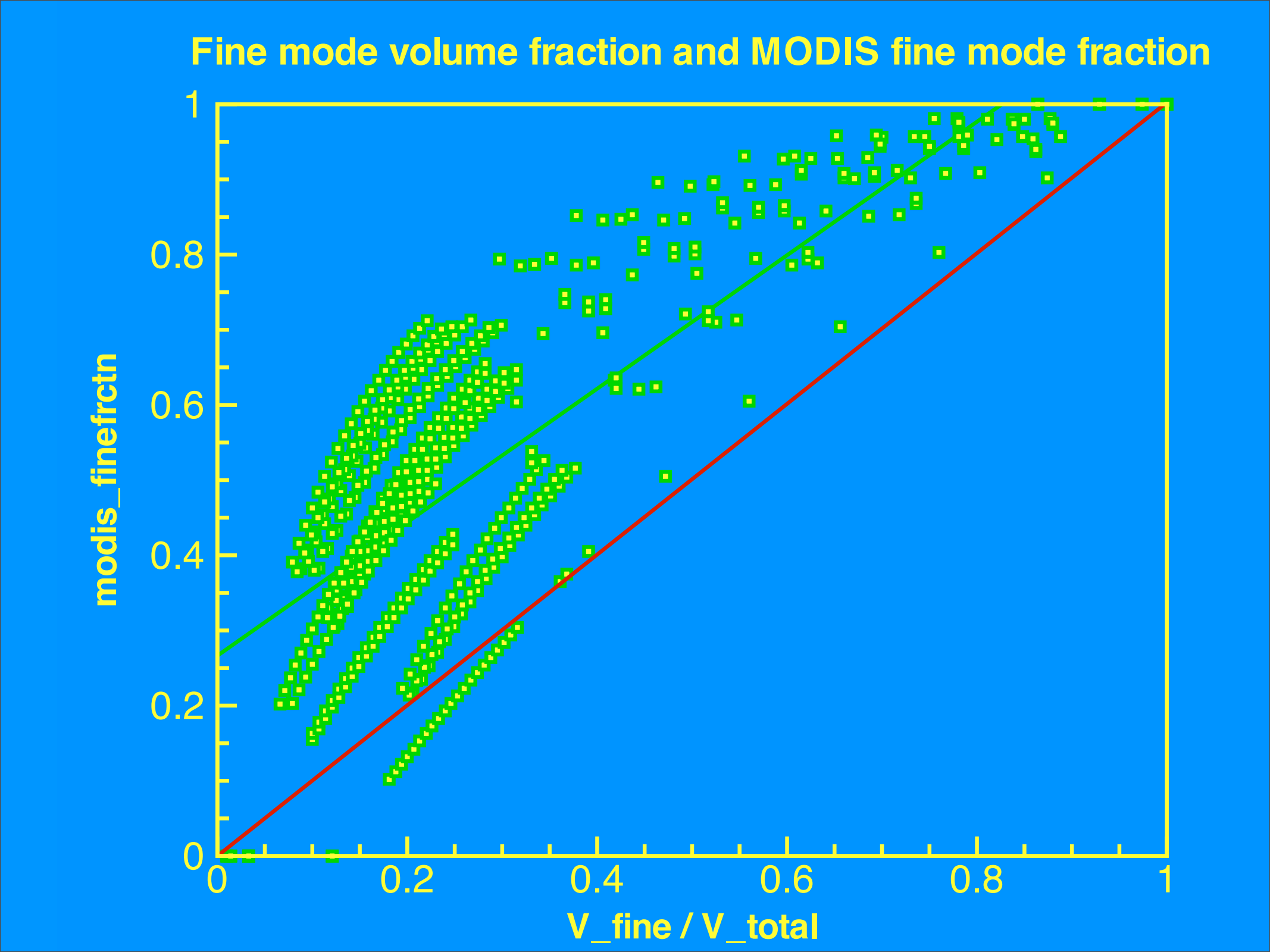
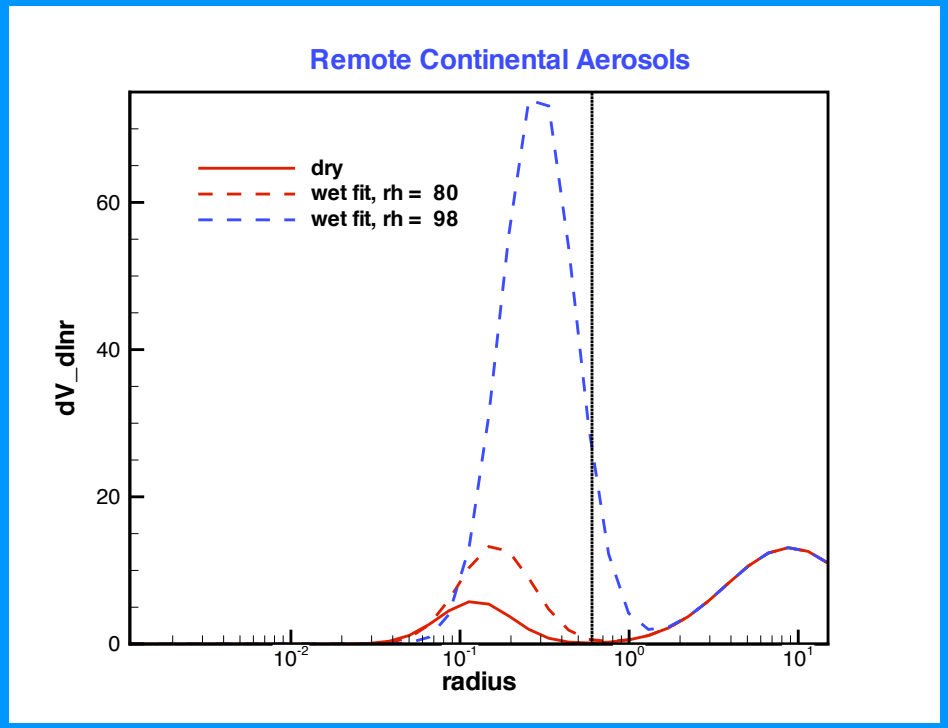

Fine mode volume fraction and MODIS fine mode fraction
modis_finefrctn
V_fine / V_total
0
0.2
0.4
0.6
0.8
1

A theoretical study of hygroscopic aerosol growth

- Dry size distributions after Jaenicke (1993) or Dubovik et al. (2002).
- Hygroscopic growth with relative humidity as parameterized by Tang and Munkelwitz (1994).
- Solute refractive indices from Tang and Munkelwitz, 1991, and Lacis, http://gacp.giss.nasa.gov/data_sets



- Optical depth and Angstrom exponent calculations via Mie Theory (Wiscombe, 1980).

Part I
A Case Study with a Single Size Distribution

$$\frac{dV}{d\ln r} = \sum_{i=1}^{N} \frac{C_i}{\sqrt{2\pi}\sigma_i} \exp\left[\frac{-(\ln r - \ln R_i)^2}{2\sigma_i^2}\right]$$

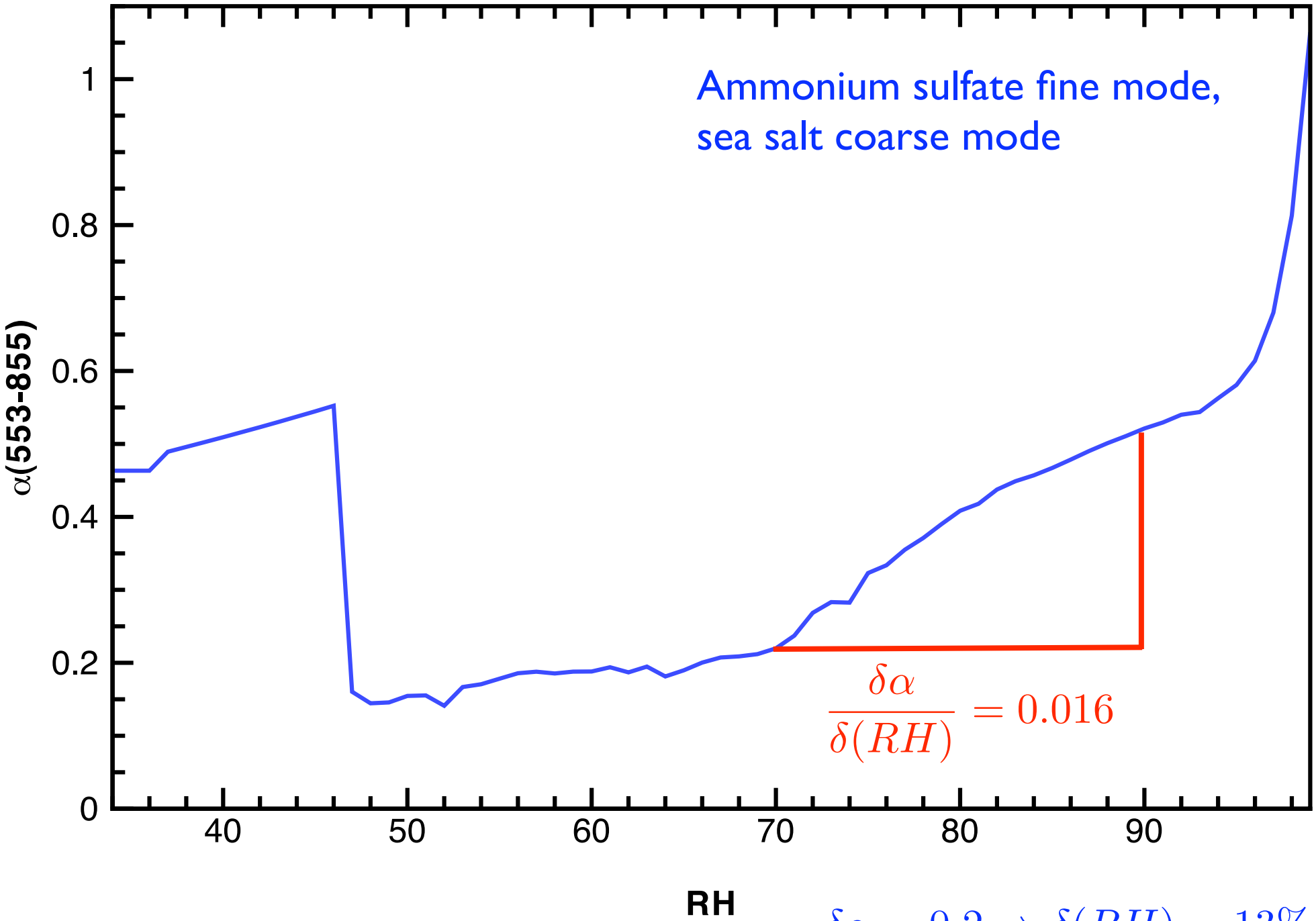
$$R_{accum} = 0.06\,\mu m$$

$$R_{crs} = 3.2\,\mu m$$

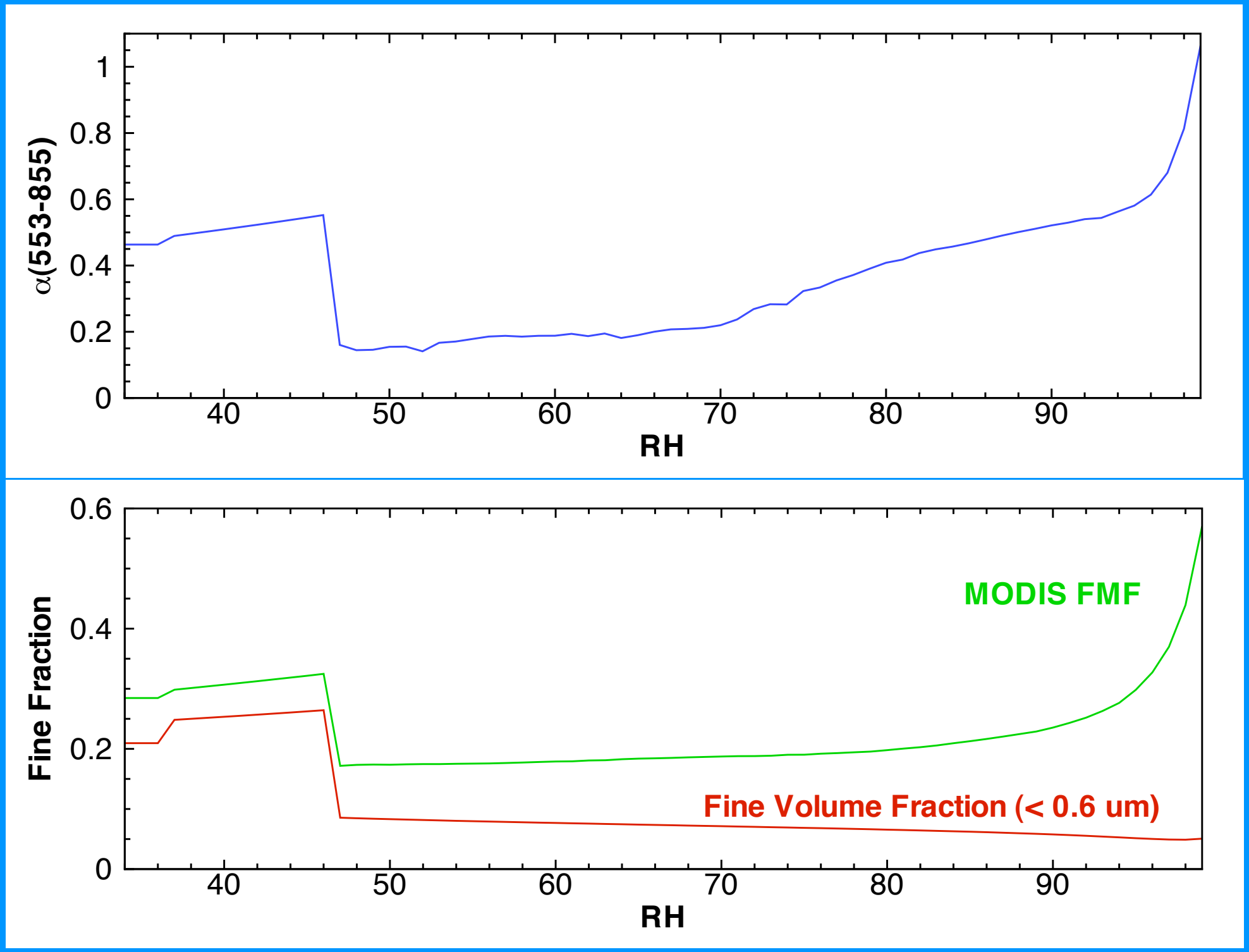
$$\sigma_{accum} = 0.38$$

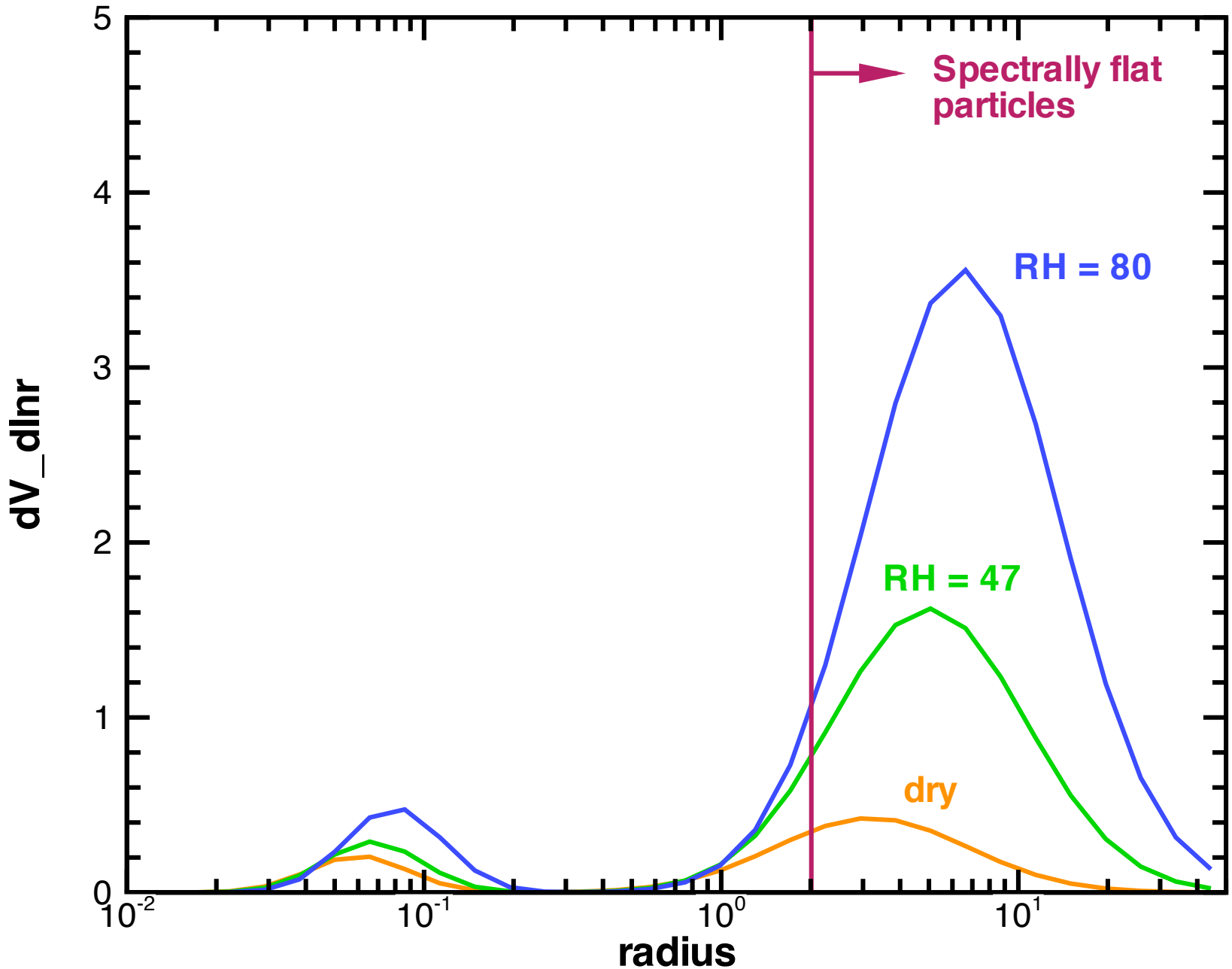
$$\sigma_{crs} = 0.75$$

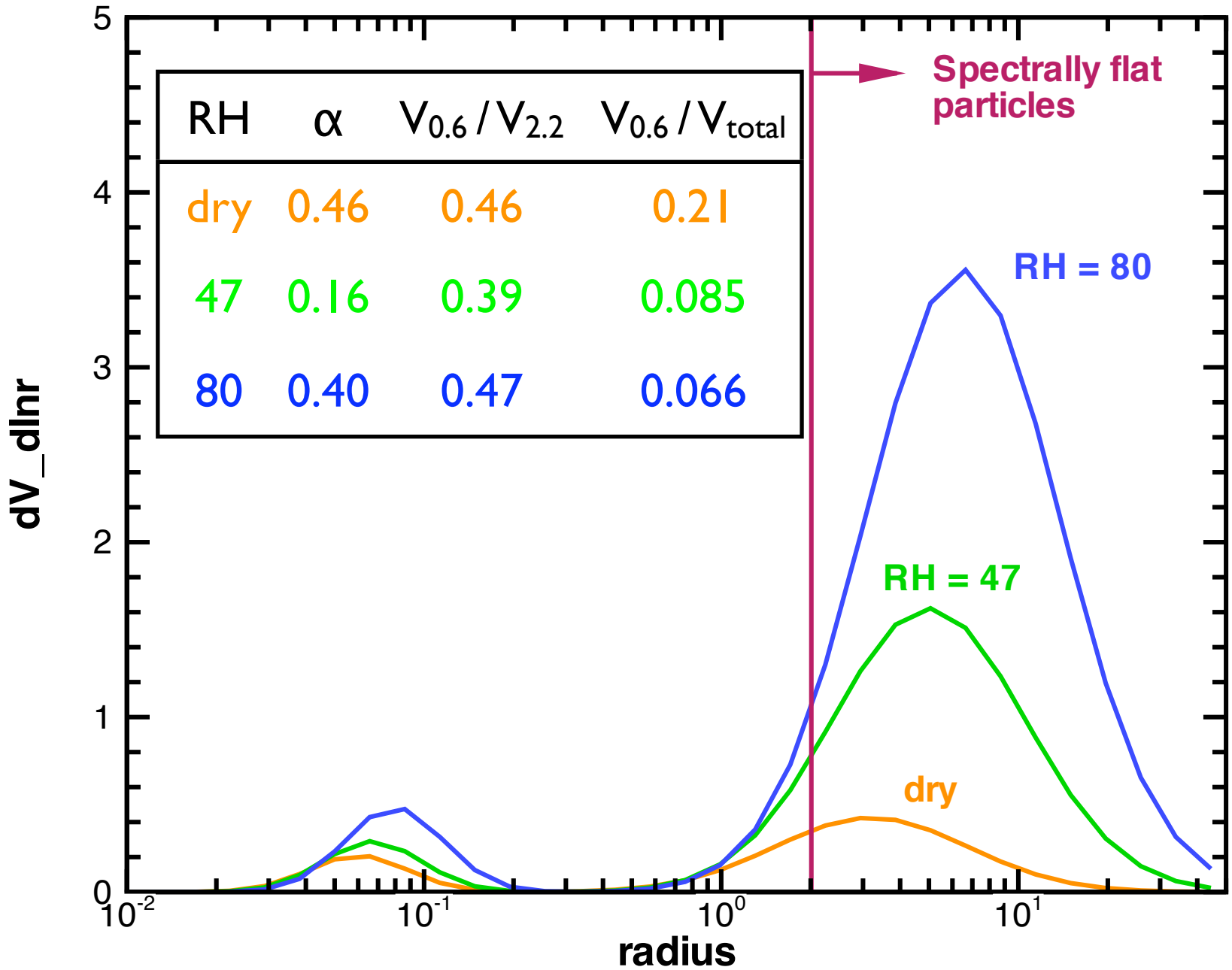
$$V_{fine}/V_{total}(dry) = 0.25$$



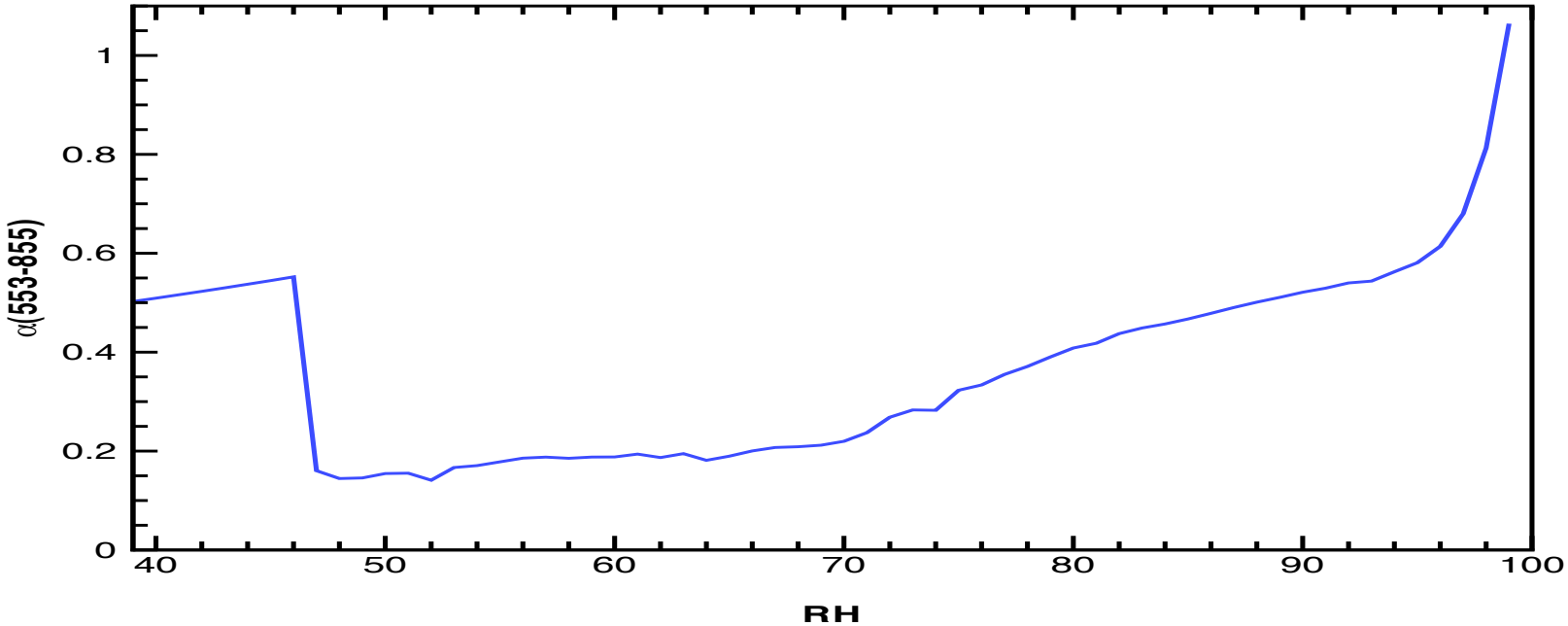
$\delta\alpha = 0.2 \Rightarrow \delta(RH) = 13\%$

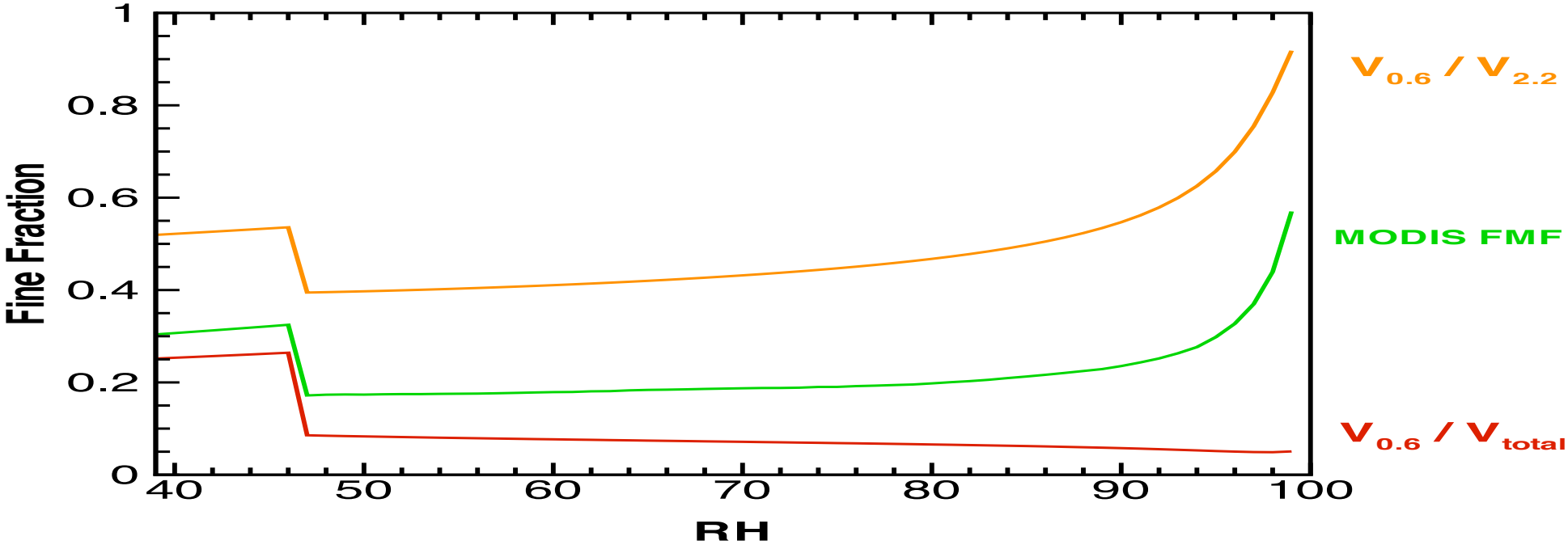

α(553-855)
1
0.8
0.6
0.4
0.2
0
40
50
60
70
80
90
RH
Fine Fraction
0.6
0.4
0.2
0
MODIS FMF
Fine Volume Fraction (< 0.6 um)
40
50
60
70
80
90
RH

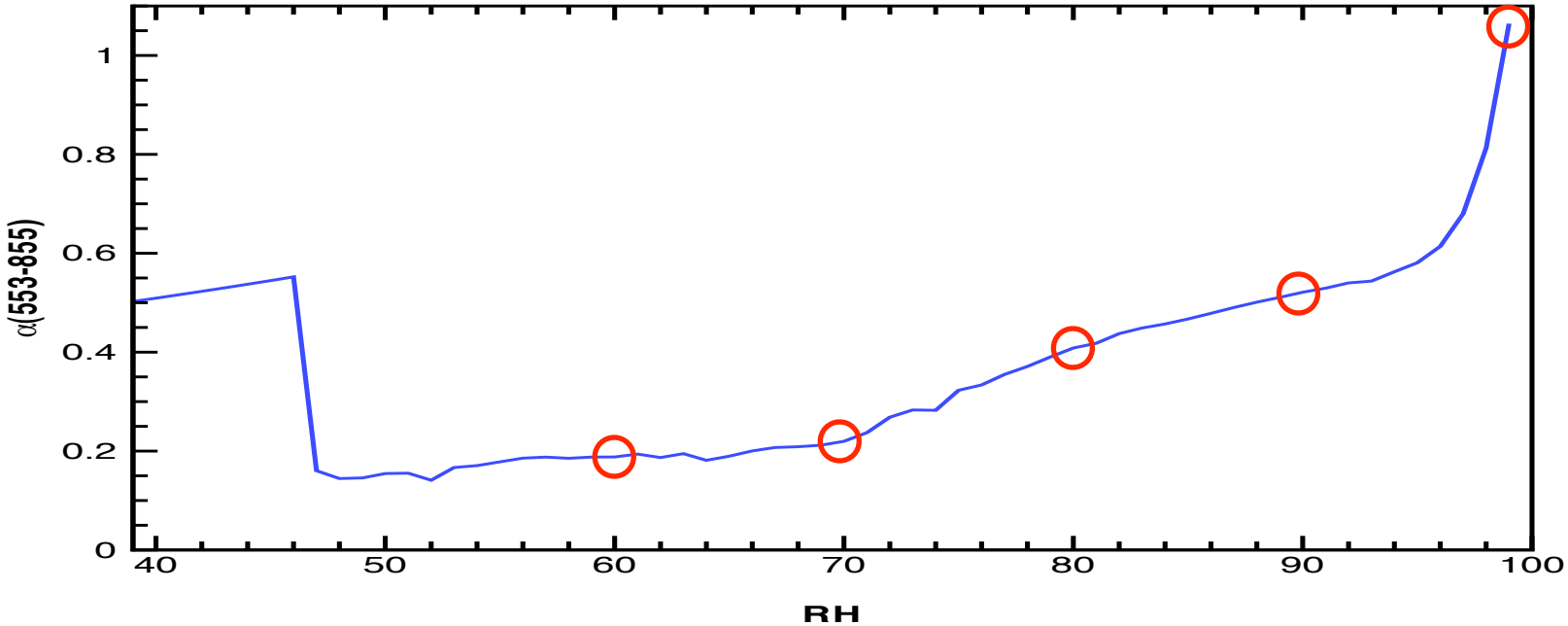

Spectrally flat particles
RH = 80
RH = 47
dry
dV_dlnr
radius
0
1
2
3
4
5
10^{-2}
10^{-1}
10^{0}
10^{1}

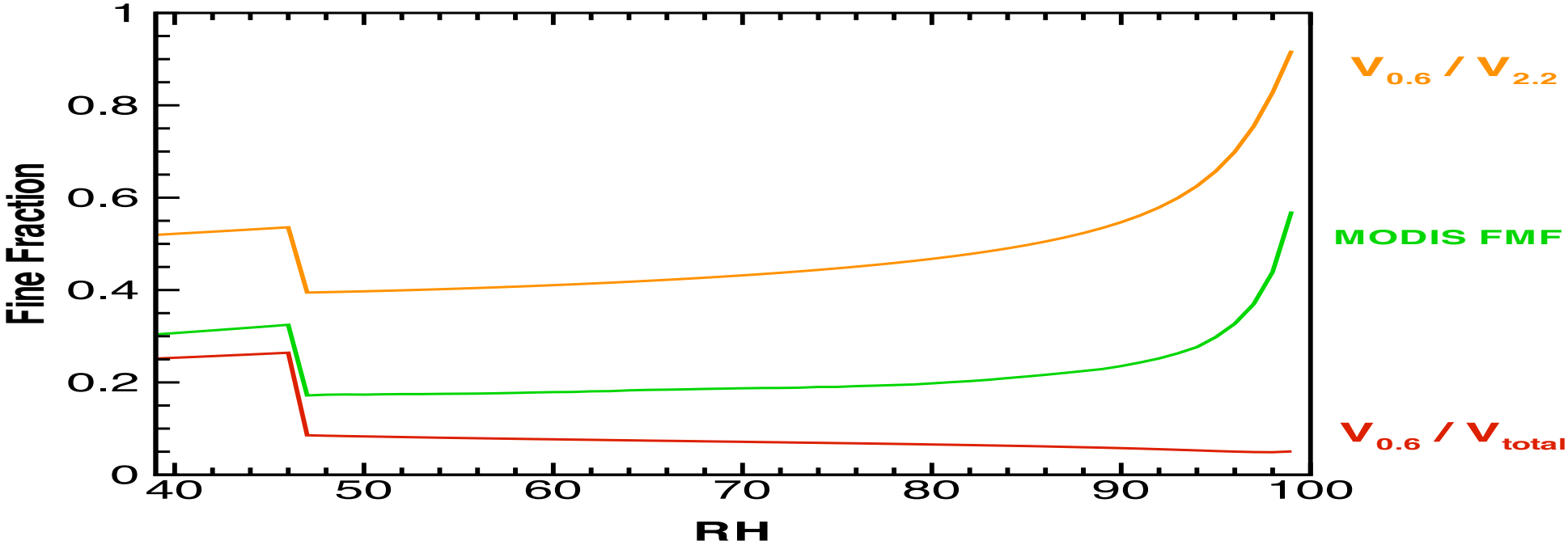


RH	α	$V_{0.6} / V_{2.2}$	$V_{0.6} / V_{total}$
dry	0.46	0.46	0.21
47	0.16	0.39	0.085
80	0.40	0.47	0.066


α(553-855)
0
0.2
0.4
0.6
0.8
1
40
50
60
70
80
90
100
RH


Fine Fraction
0
0.2
0.4
0.6
0.8
1
40
50
60
70
80
90
100
RH
V0.6 / V2.2
MODIS FMF
V0.6 / Vtotal


α(553-855)
0
0.2
0.4
0.6
0.8
1
40
50
60
70
80
90
100
RH

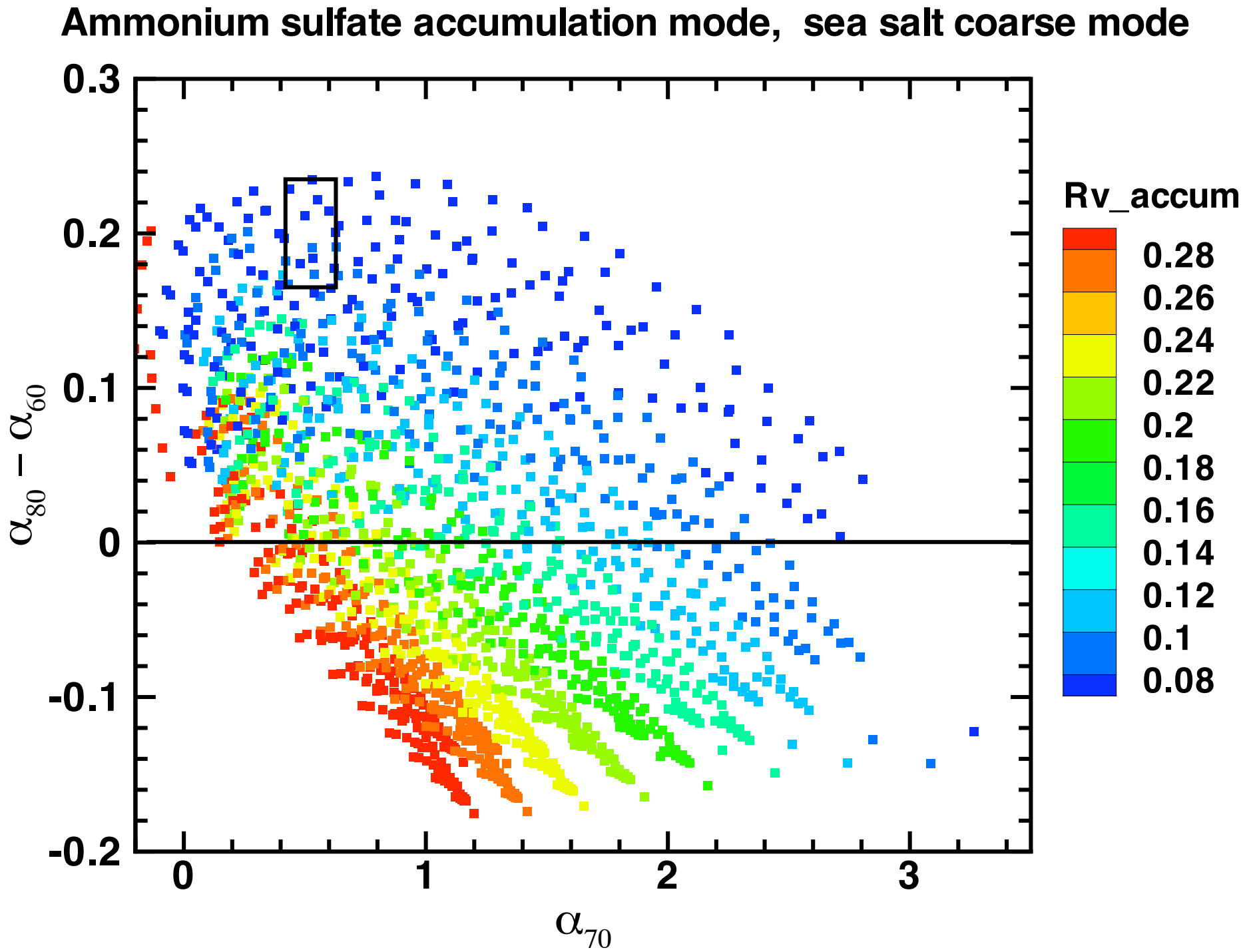

Fine Fraction
0
0.2
0.4
0.6
0.8
1
40
50
60
70
80
90
100
RH
V0.6 / V2.2
MODIS FMF
V0.6 / Vtotal

Part II
More Testing --1980 dry size distributions

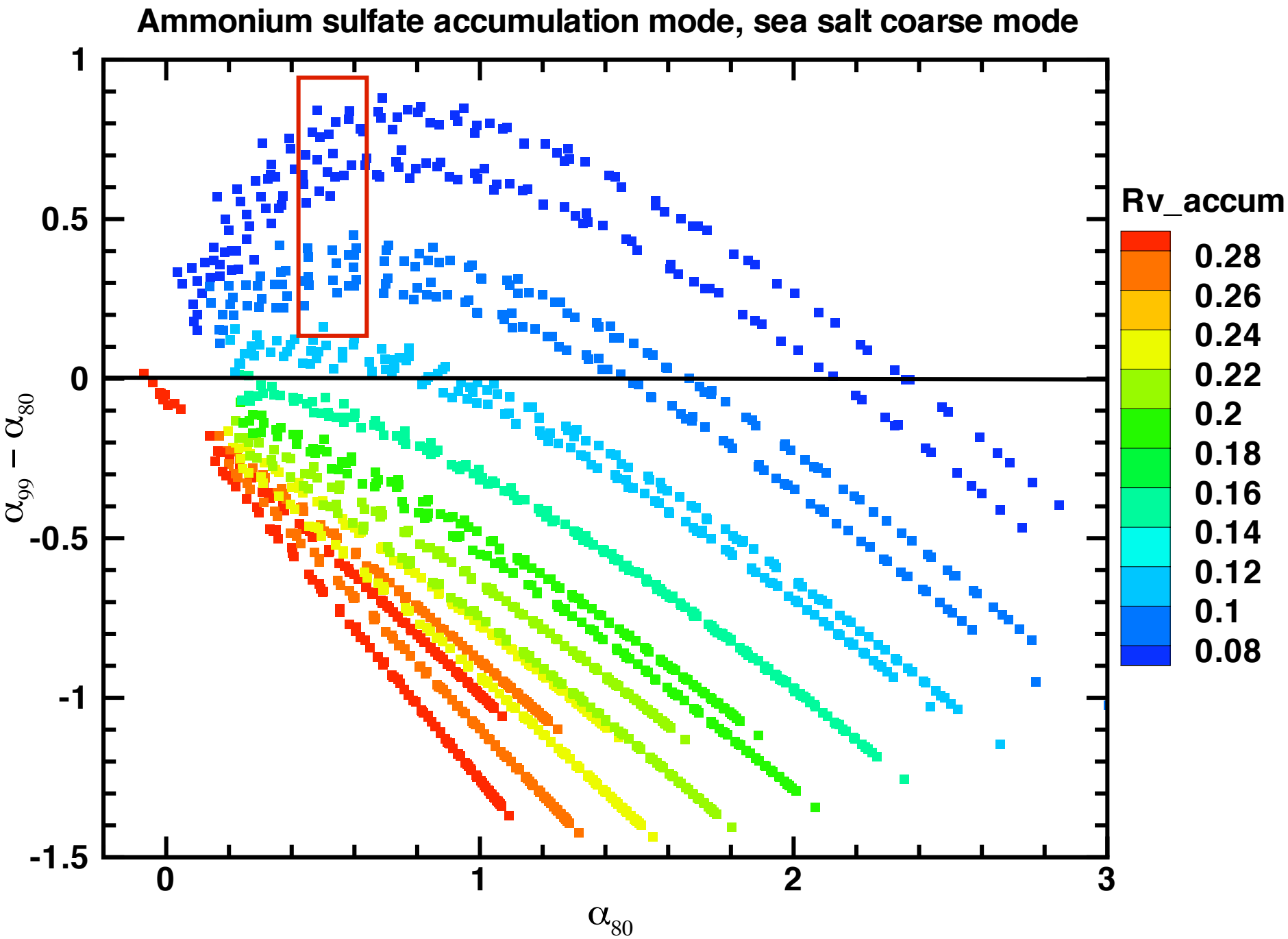
$$\frac{dV}{d\ln r} = \sum_{i=1}^{N} \frac{C_i}{\sqrt{2\pi}\sigma_i} \exp\left[\frac{-(\ln r - \ln R_i)^2}{2\sigma_i^2}\right]$$

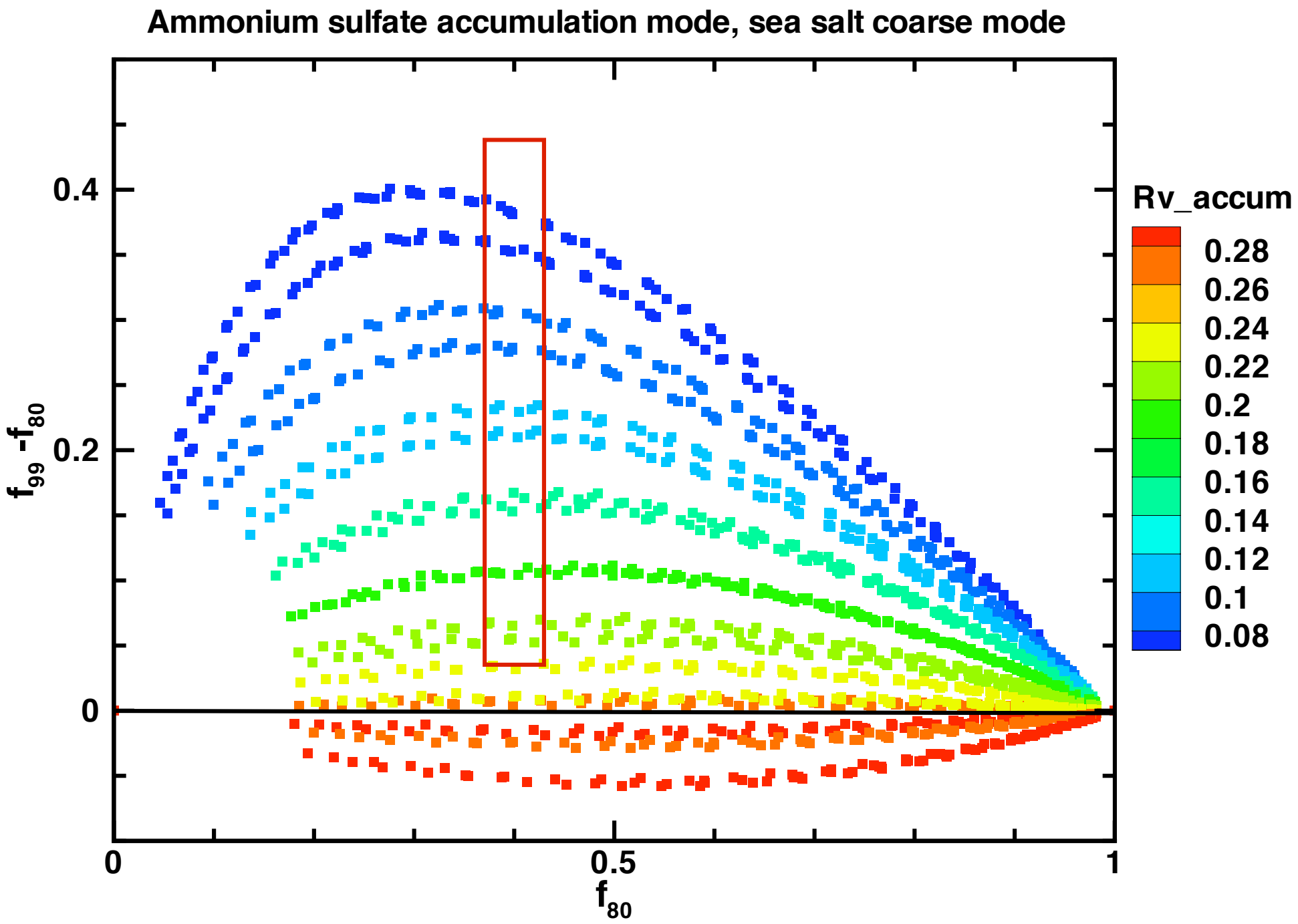
Dubovik (2002) climatology:

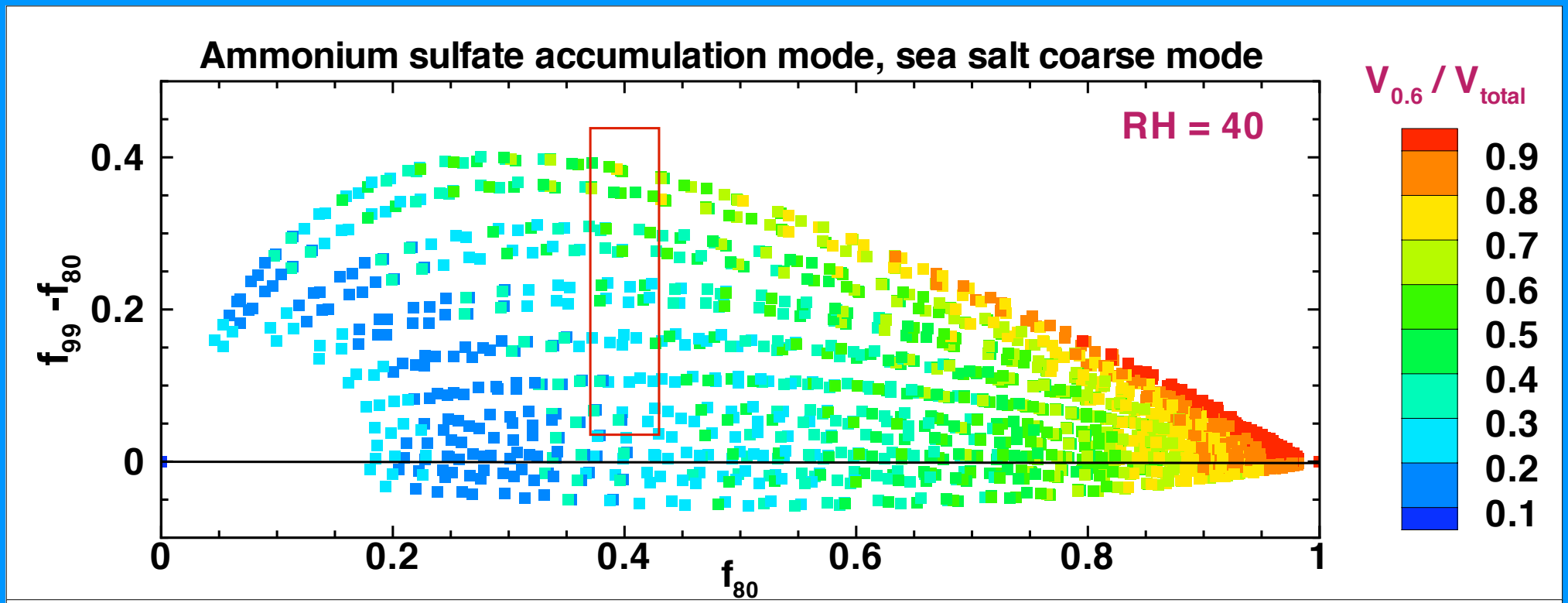
Parameter	**Value**
$R_{fine}(\mu\mathrm{m})$	**0.06, 0.09, 0.12, 0.15, 0.18, 0.21, 0.24, 0.27, 0.30**
σ_{fine}	**0.38**, 0.5
$R_{crs}(\mu\mathrm{m})$	1.9, 2.2, 2.7, **3.2**, 3.7
σ_{crs}	**0.75**, 1.0
C_{fine}/C_{total}	0.0, **0.1**, 0.2, **0.3**, 0.4, 0.5, **0.6**, 0.7, **0.8**, 0.9, **1.0**

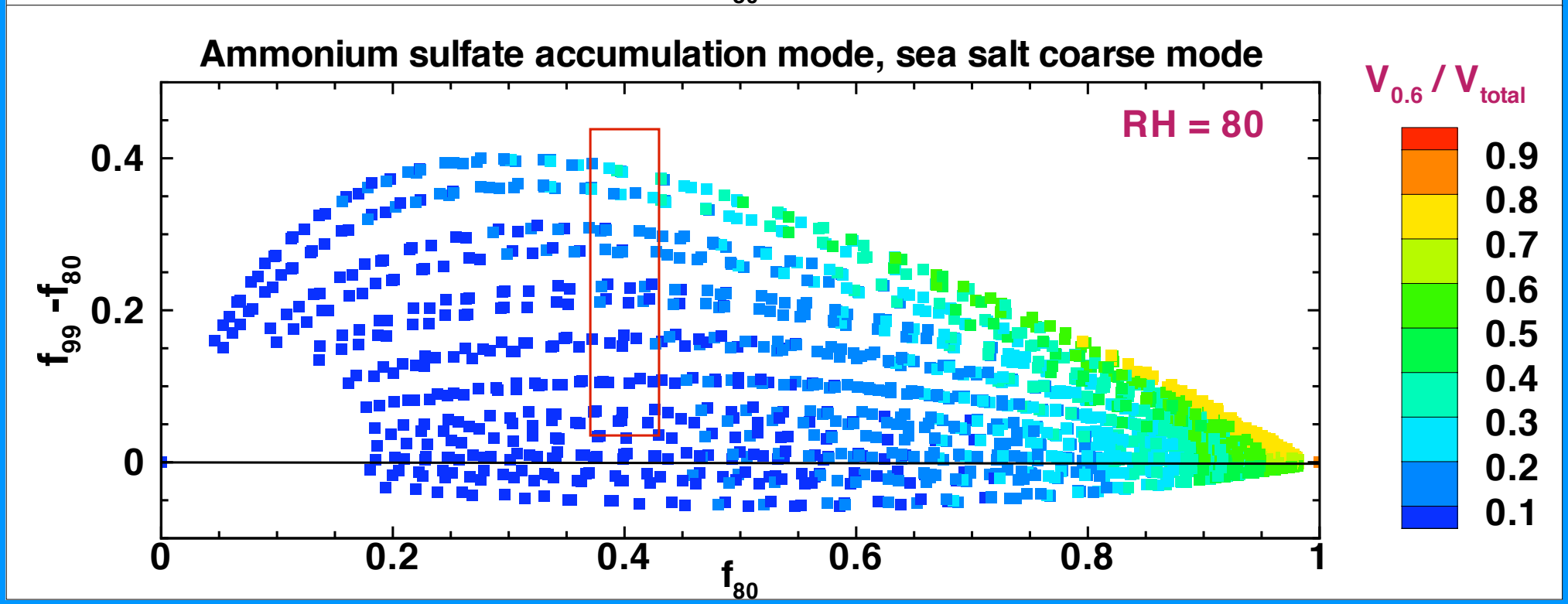

Ammonium sulfate accumulation mode, sea salt coarse mode
0.3
0.2
0.1
0
-0.1
-0.2
0
1
2
3
$\alpha_{80} - \alpha_{60}$
α_{70}
Rv_accum
0.28
0.26
0.24
0.22
0.2
0.18
0.16
0.14
0.12
0.1
0.08

What happens if we get closer to clouds?


Ammonium sulfate accumulation mode, sea salt coarse mode
1
0.5
0
-0.5
-1
-1.5
0
1
2
3
$\alpha_{99} - \alpha_{80}$
α_{80}
Rv_accum
0.28
0.26
0.24
0.22
0.2
0.18
0.16
0.14
0.12
0.1
0.08

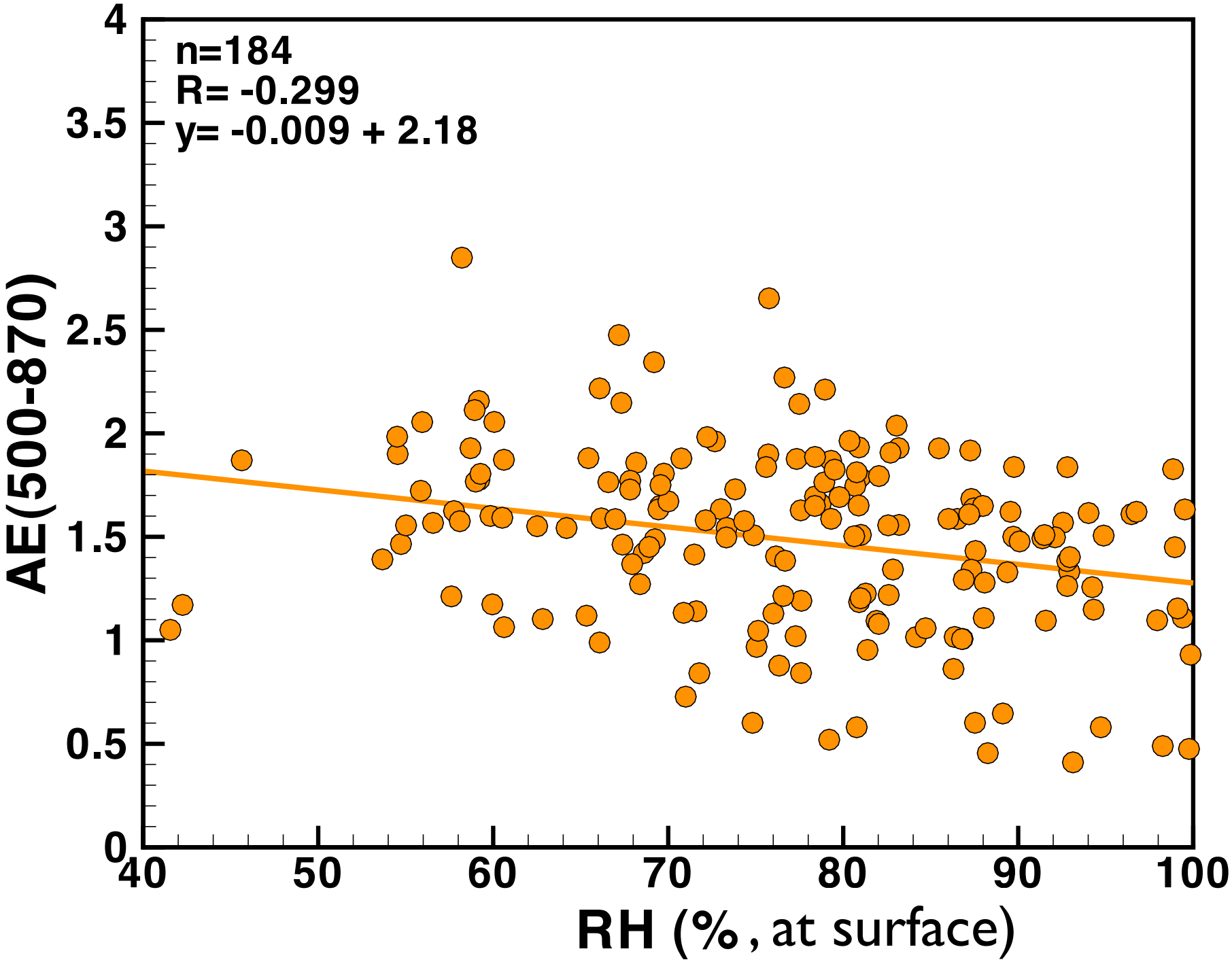

Ammonium sulfate accumulation mode, sea salt coarse mode
0.4
0.2
0
f_{99} -f_{80}
0
0.5
1
f_{80}
Rv_accum
0.28
0.26
0.24
0.22
0.2
0.18
0.16
0.14
0.12
0.1
0.08

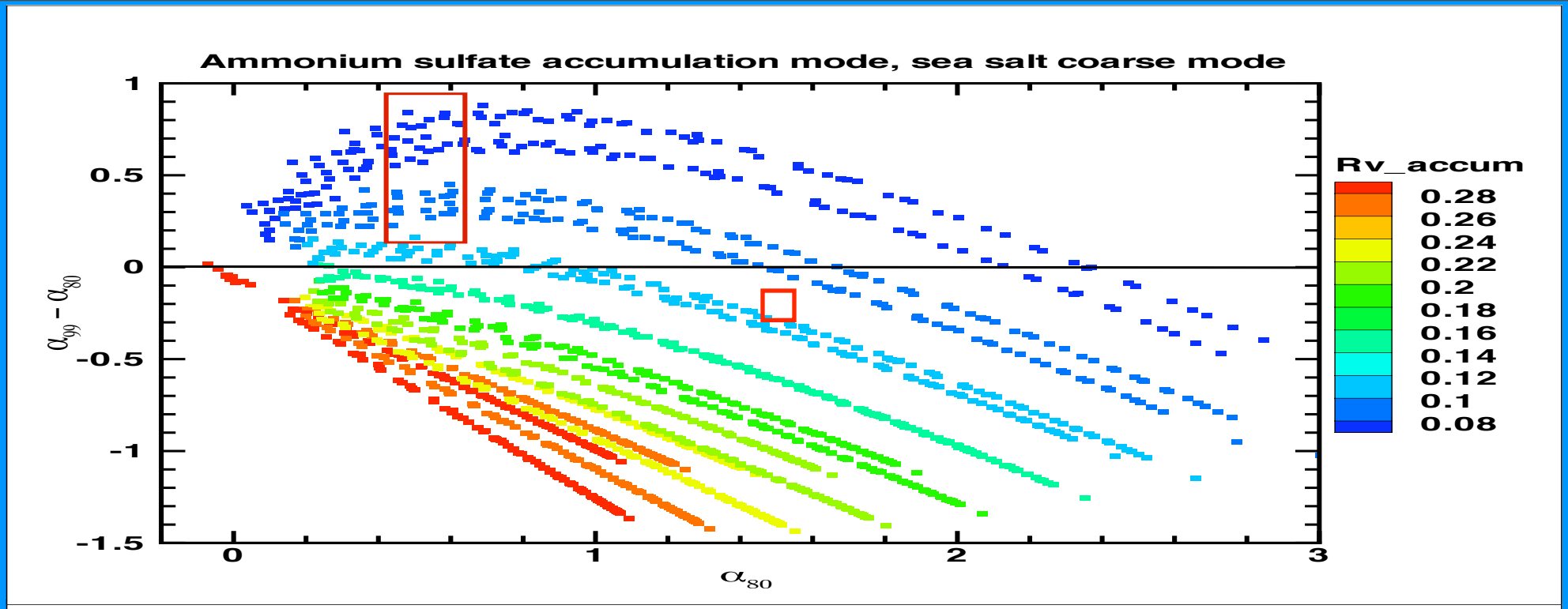

Ammonium sulfate accumulation mode, sea salt coarse mode
RH = 40
$V_{0.6} / V_{total}$
0.9
0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1
$f_{99} - f_{80}$
0.4
0.2
0
0
0.2
0.4
0.6
0.8
1
f_{80}

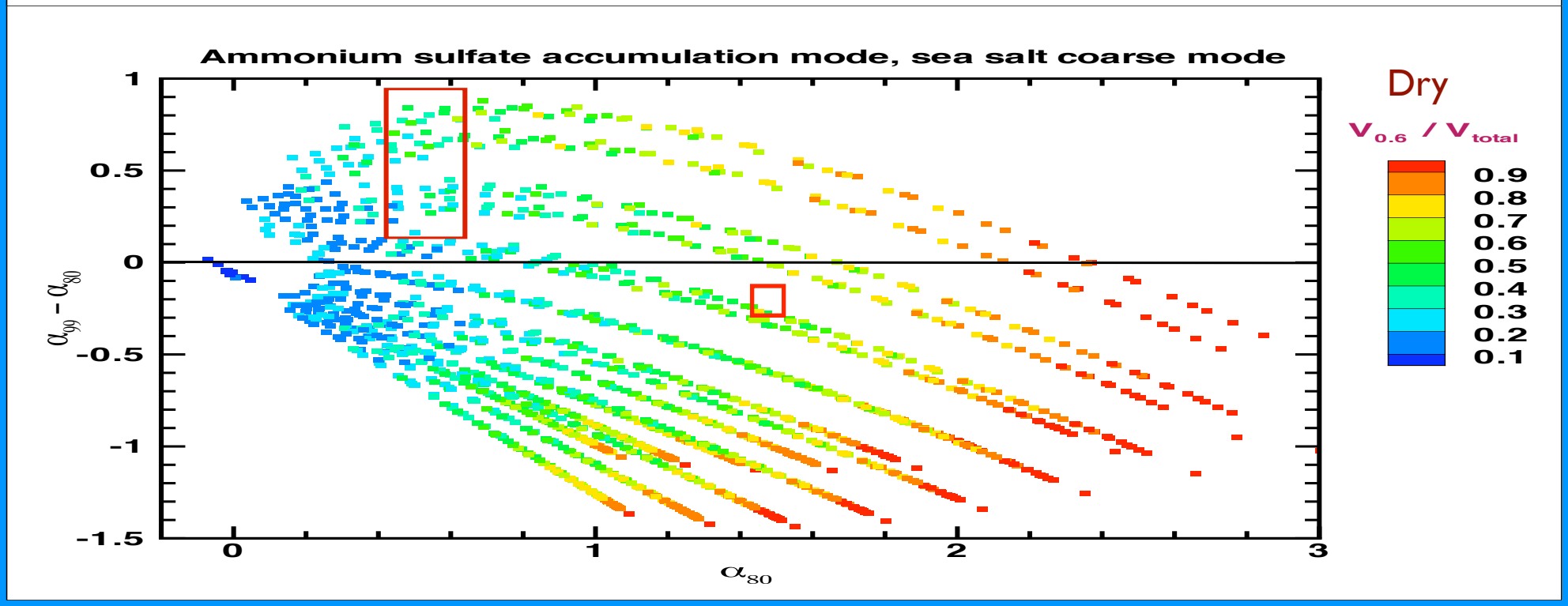

Ammonium sulfate accumulation mode, sea salt coarse mode
RH = 80
$V_{0.6} / V_{total}$
0.9
0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1
$f_{99} - f_{80}$
0.4
0.2
0
0
0.2
0.4
0.6
0.8
1
f_{80}

Preliminary Ground Data from COVE

2001-2005 RH vs AE(500-870) by Fall season with statistics




Ammonium sulfate accumulation mode, sea salt coarse mode
Rv_accum
0.28
0.26
0.24
0.22
0.2
0.18
0.16
0.14
0.12
0.1
0.08
$\alpha_{99} - \alpha_{80}$
α_{80}


Ammonium sulfate accumulation mode, sea salt coarse mode
Dry
$V_{0.6}$ / V_{total}
0.9
0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1
$\alpha_{99} - \alpha_{80}$
α_{80}

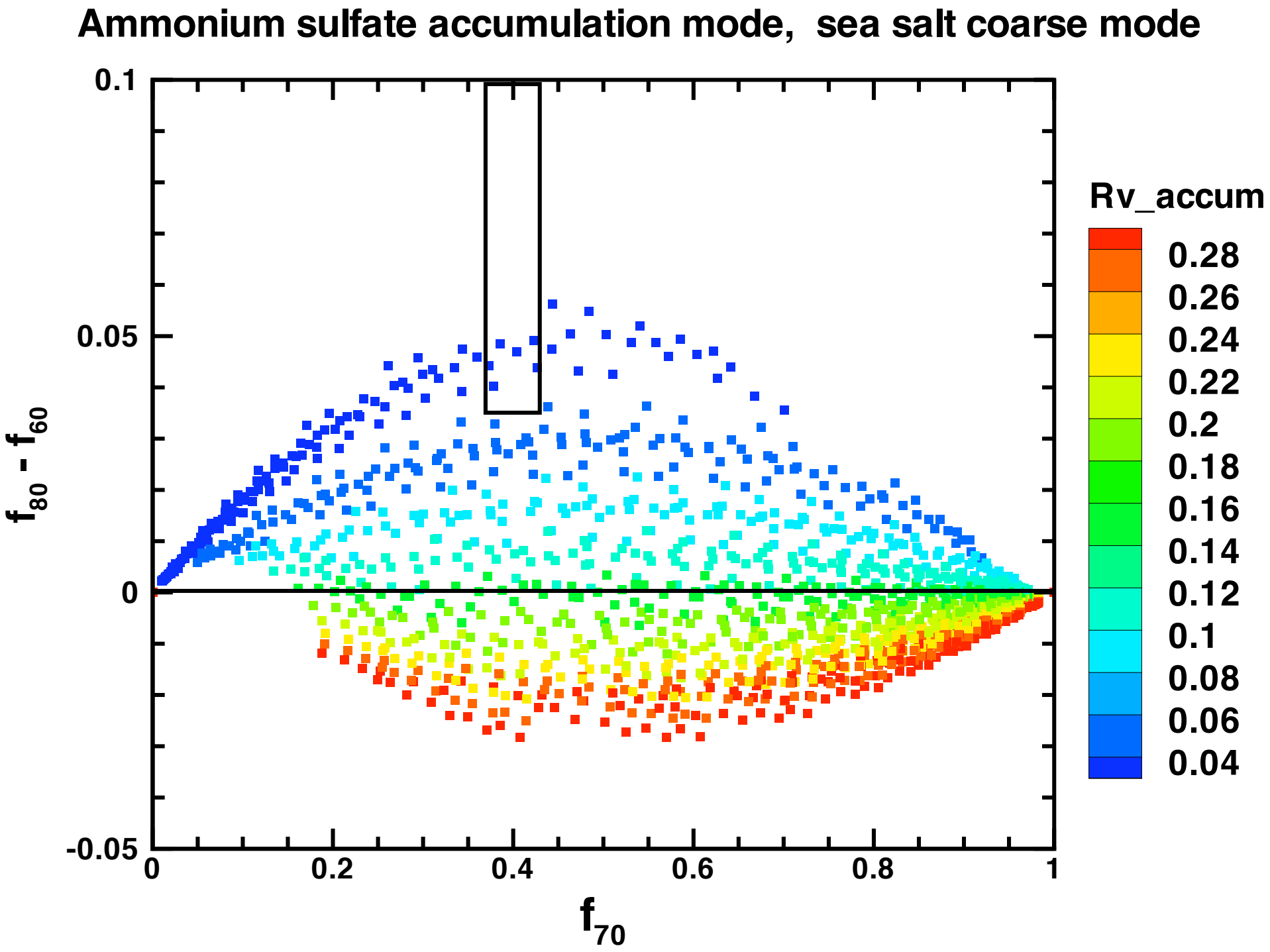
Conclusions

- Aerosol swelling *could* be responsible for the Angstrom exponent and FMF changes observed in satellite datasets.
- Ground site observations indicate an opposite Angstrom exponent effect (i.e., decreasing wrt RH), but still consistent with an aerosol swelling scenario.
- The relationship between Angstrom exponents, MODIS fine mode fractions, and aerosol fine mode volume fraction is not rigorous.

"...you keep using that word. I do not think that it means what you think it means."

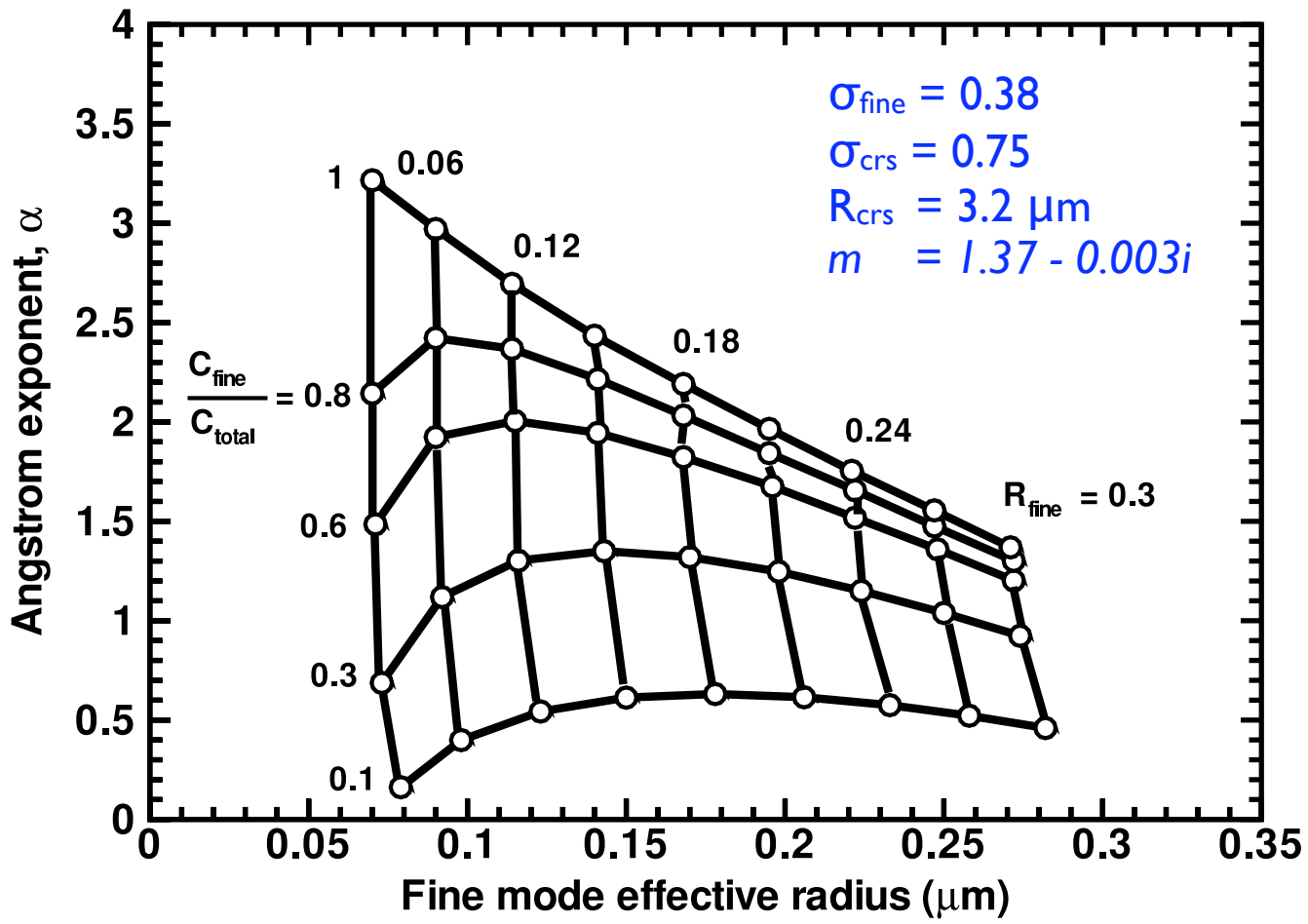
-- Inigo Montoya, in *The Princess Bride*

Appendix


Ammonium sulfate accumulation mode, sea salt coarse mode
0.1
0.05
0
-0.05
0
0.2
0.4
0.6
0.8
1
$f_{80} - f_{60}$
f_{70}
Rv_accum
0.28
0.26
0.24
0.22
0.2
0.18
0.16
0.14
0.12
0.1
0.08
0.06
0.04

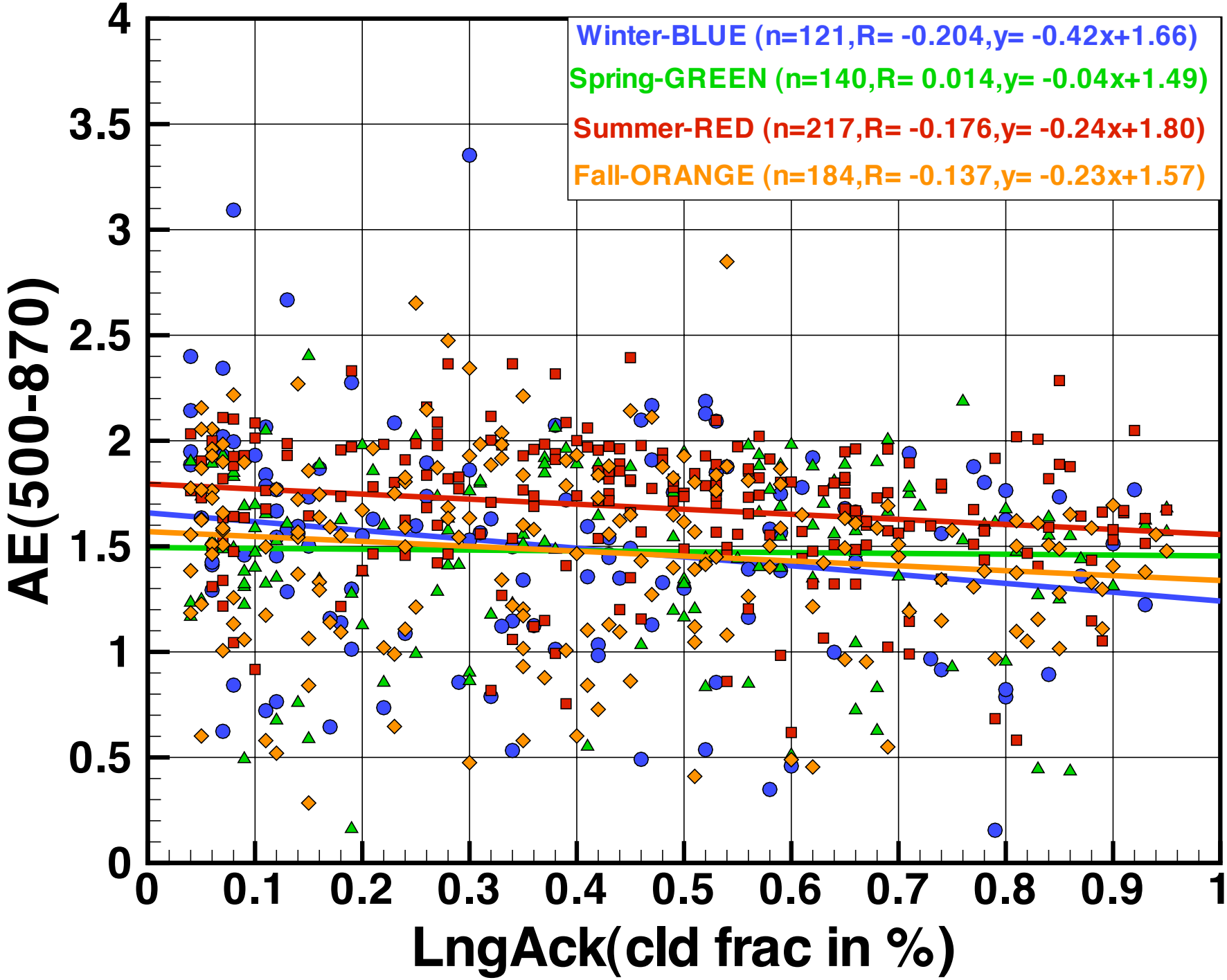
Sensitivity of Angstrom exponent to fine mode fraction and fine mode effective radius

$$\frac{dV}{d\ln r} = \sum_{i=1}^{N} \frac{C_i}{\sqrt{2\pi}\sigma_i} \exp\left[\frac{-(\ln r - \ln R_i)^2}{2\sigma_i^2}\right]$$

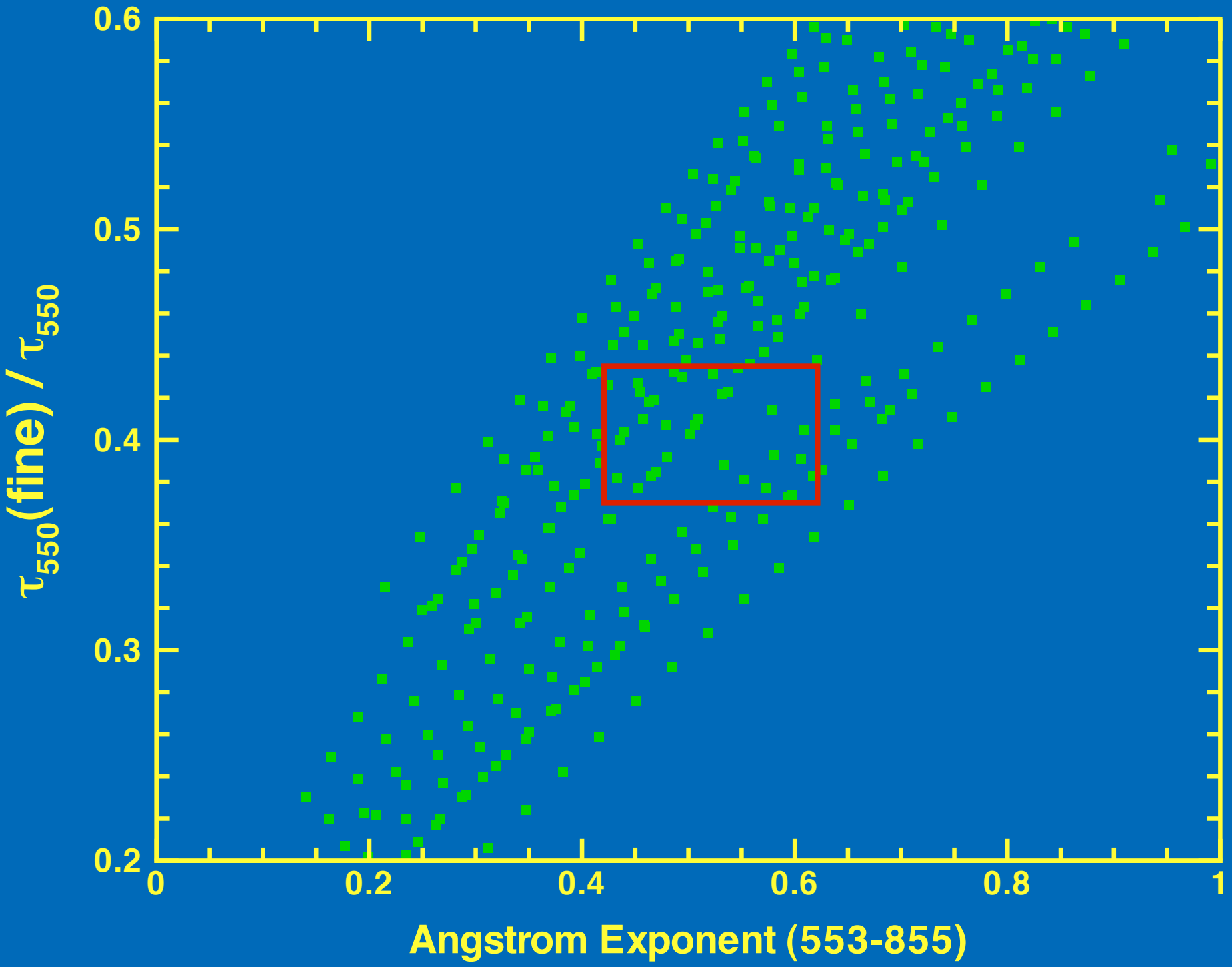


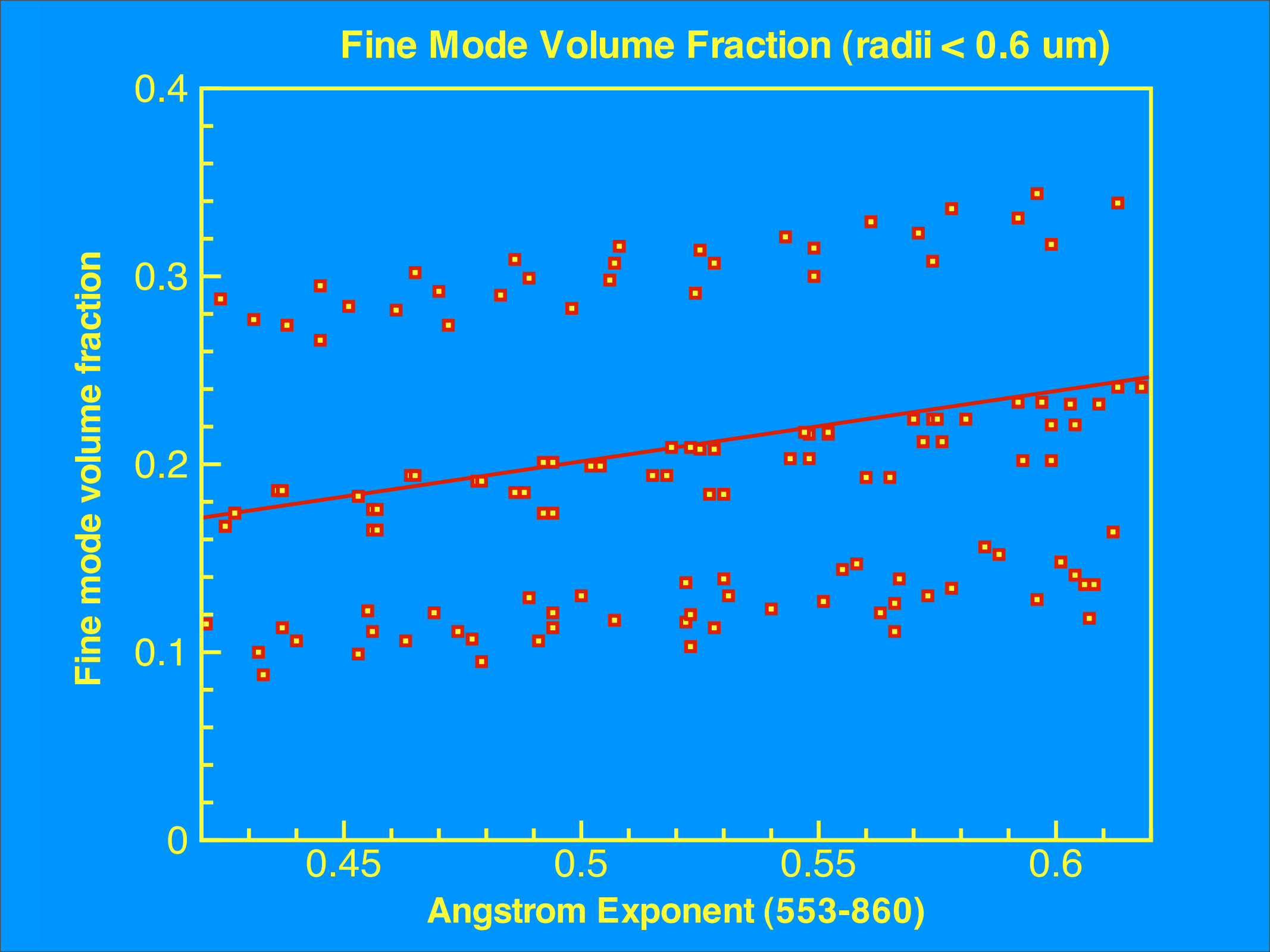
Schuster et al., JGR 2006

2001-2005 Long-Ack Cloud Fraction vs AE(500-870) by season with statistics

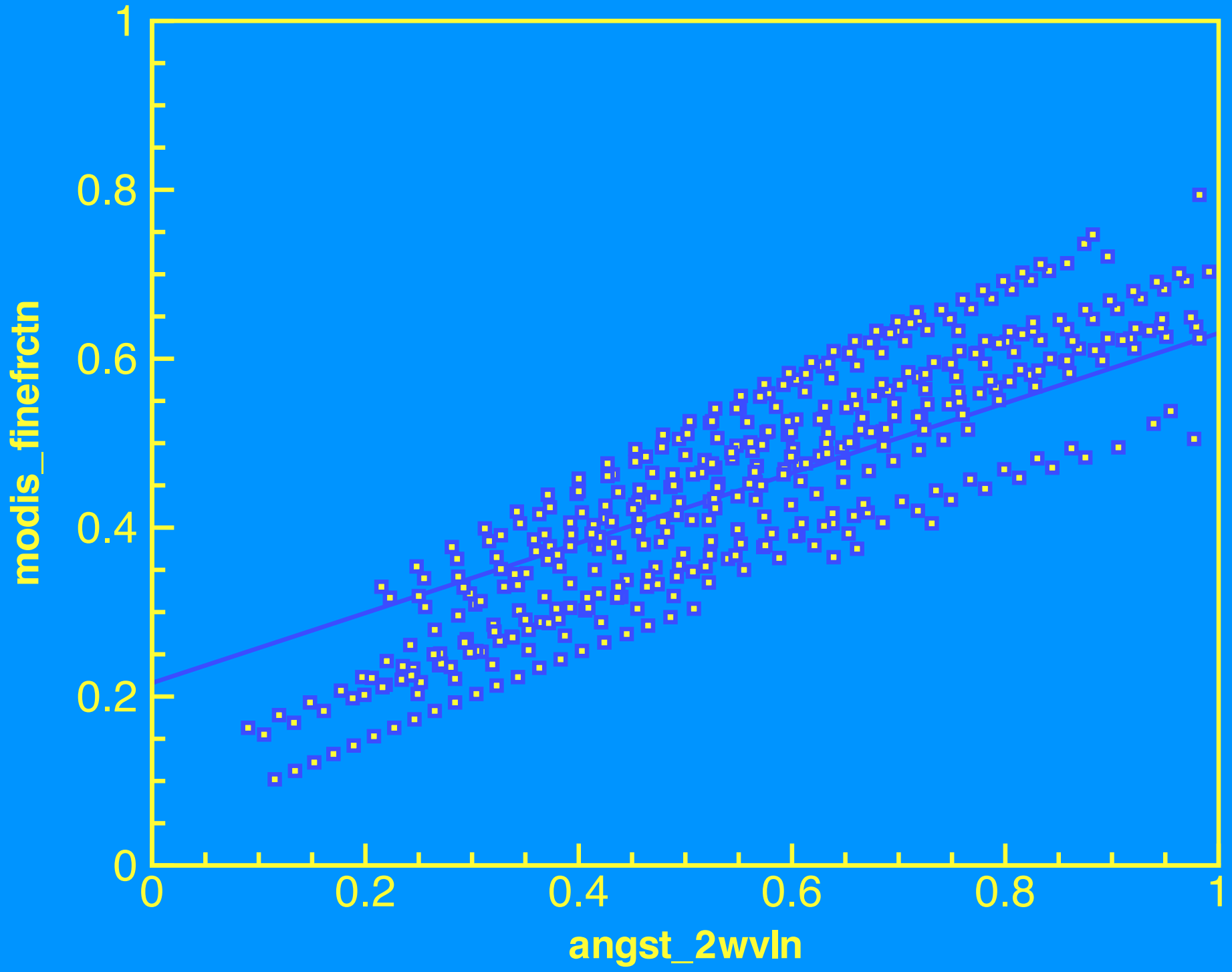


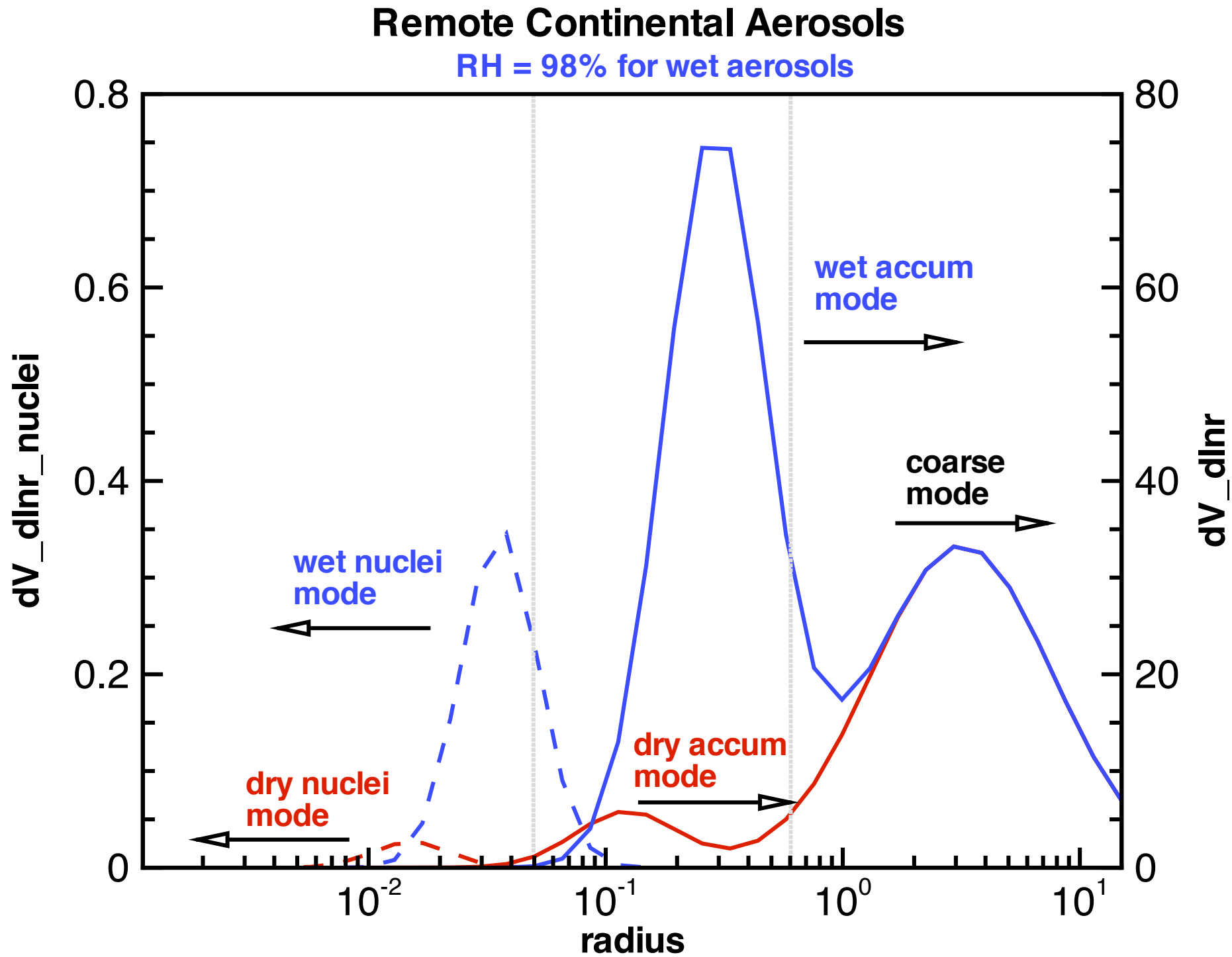
MODIS Fine Mode Fraction

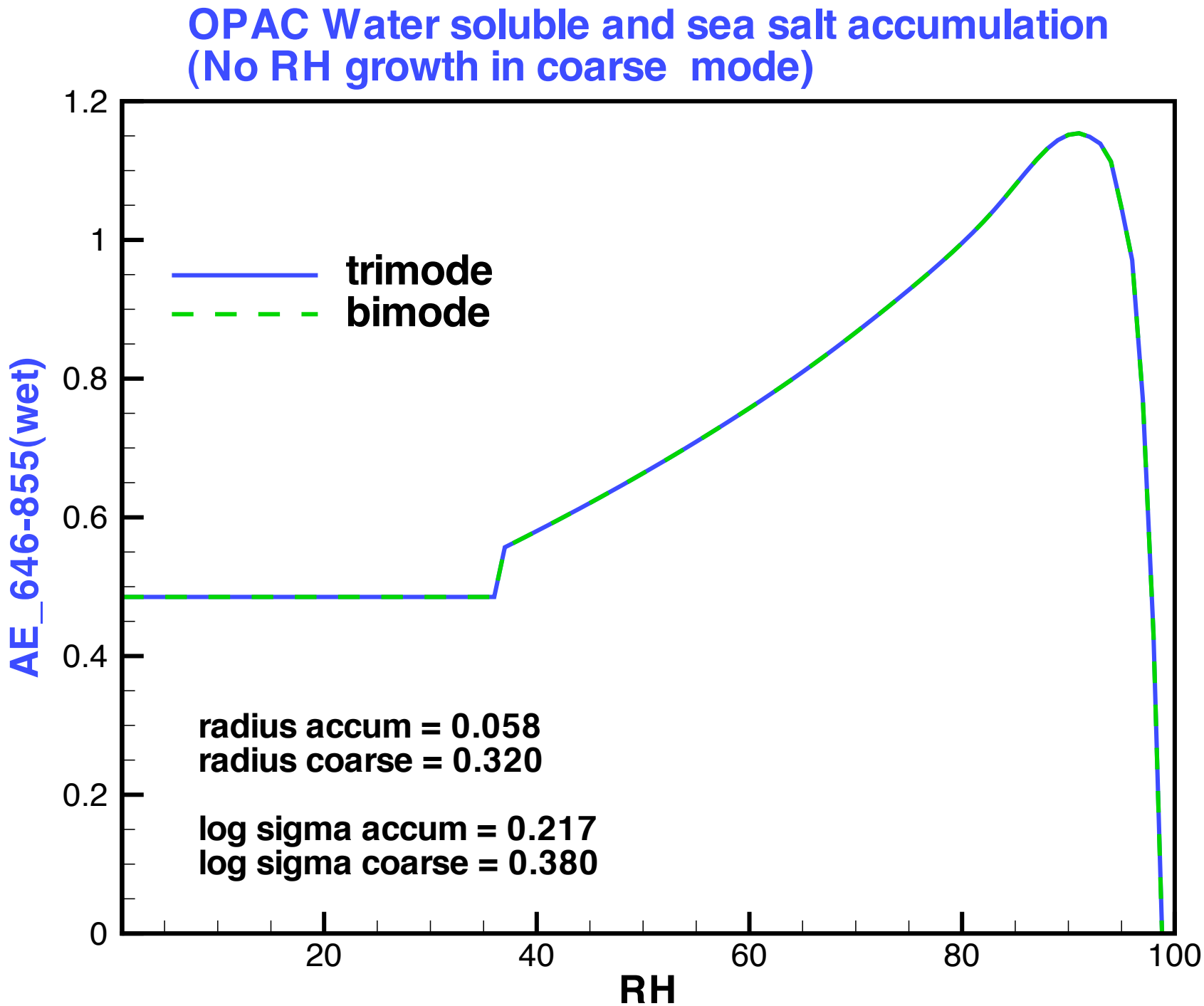


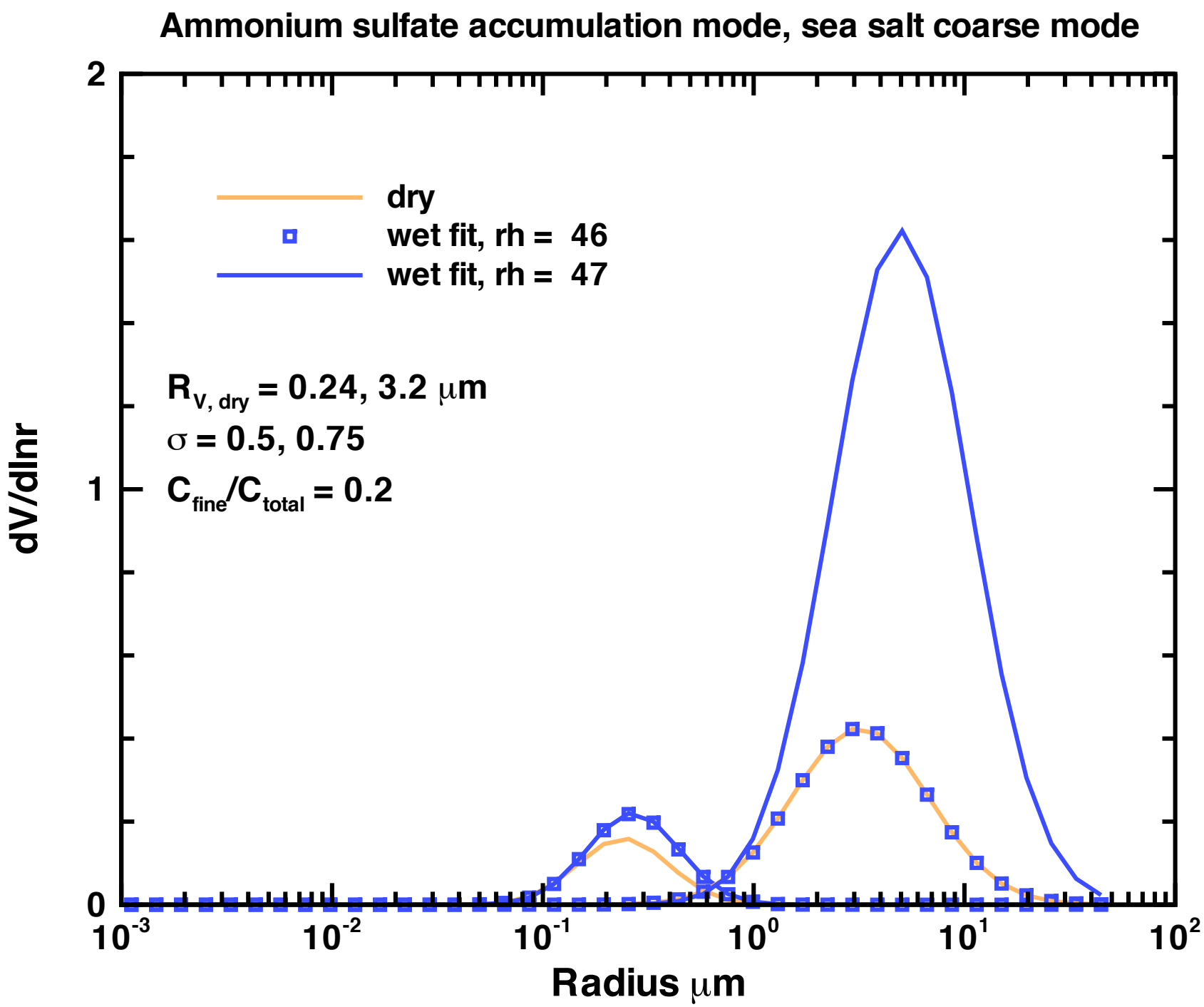

Fine Mode Volume Fraction (radii < 0.6 um)
Fine mode volume fraction
0.4
0.3
0.2
0.1
0
0.45
0.5
0.55
0.6
Angstrom Exponent (553-860)

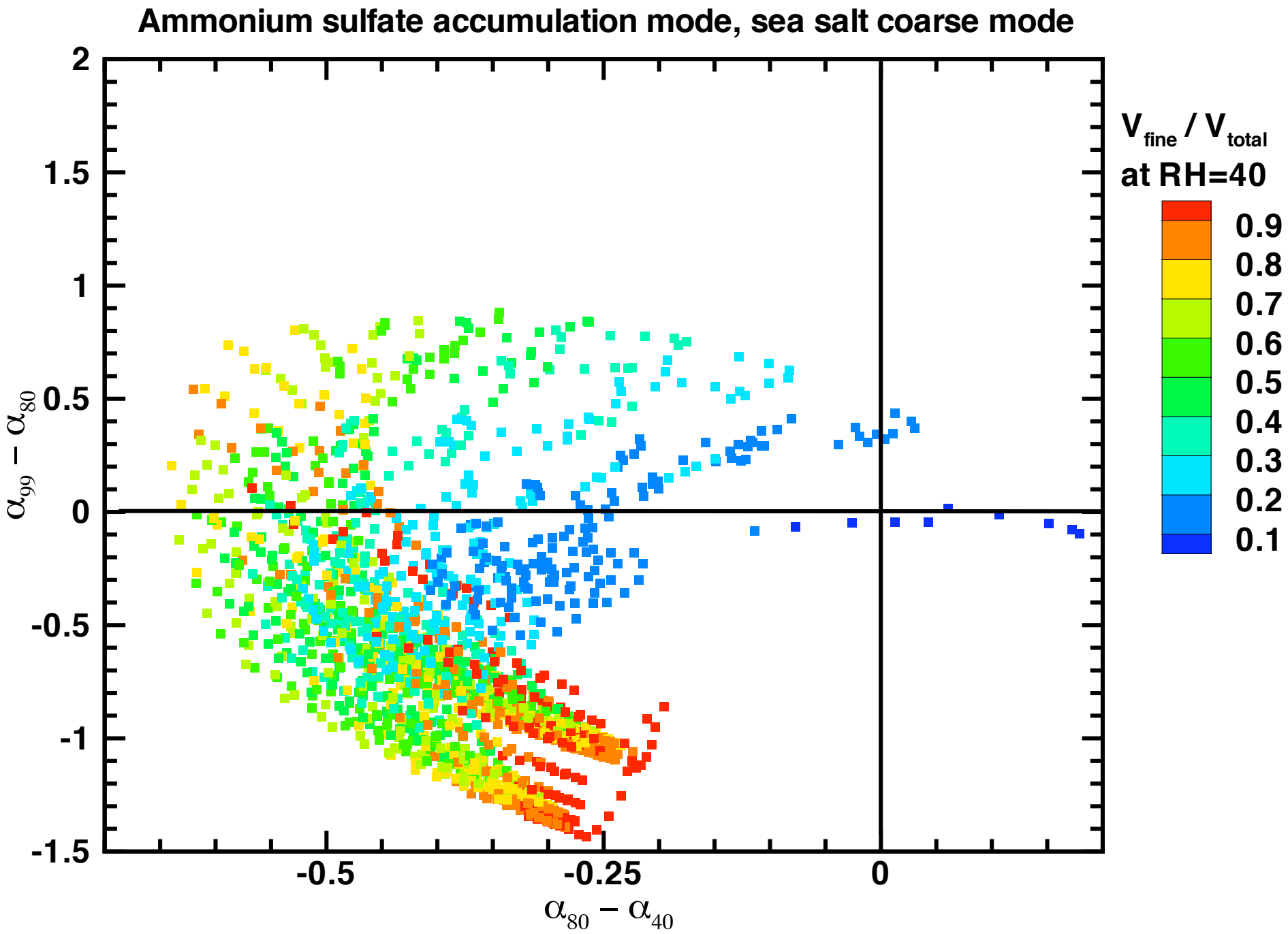
Fine mode volume fraction and MODIS fine mode fraction

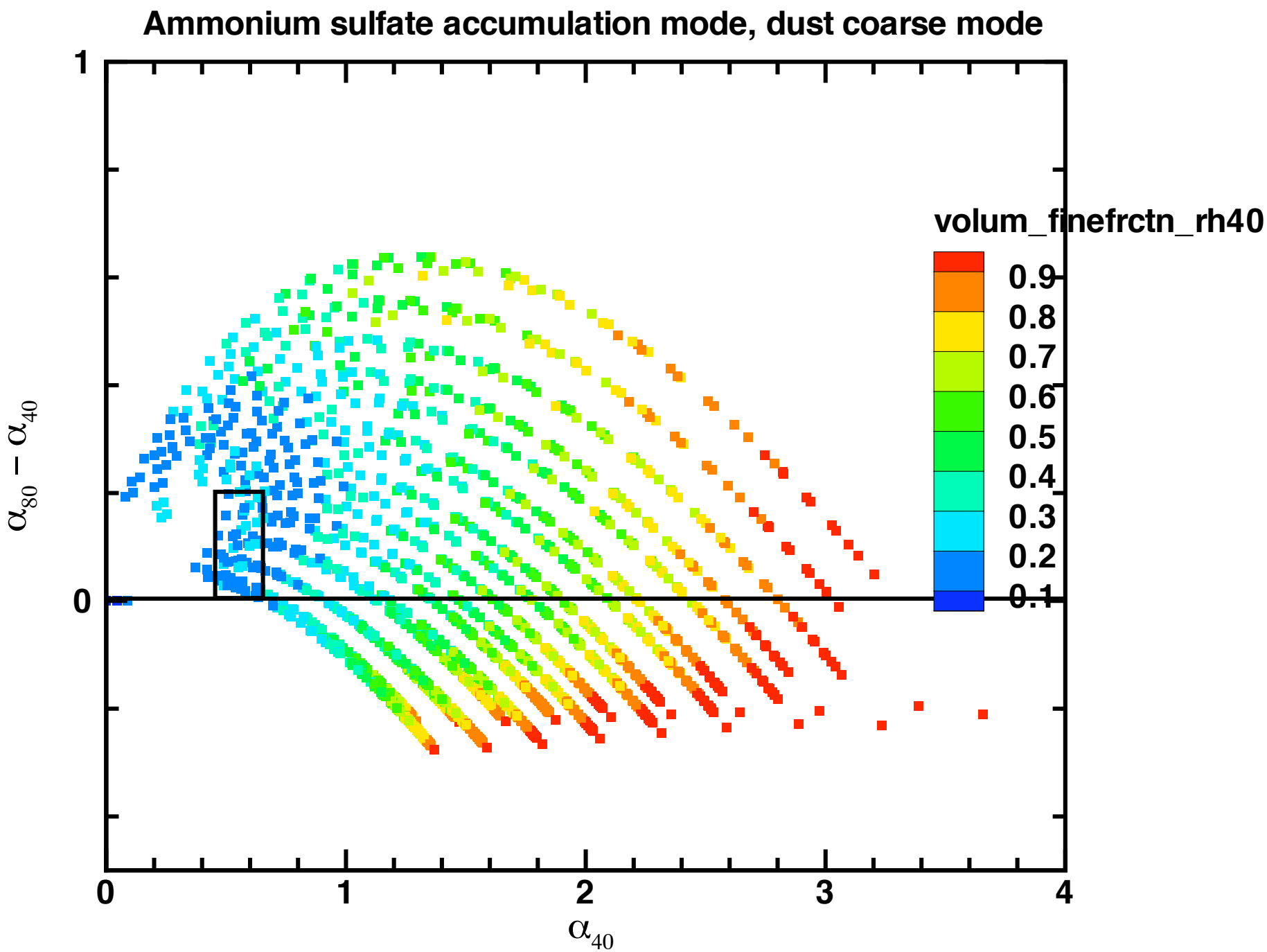


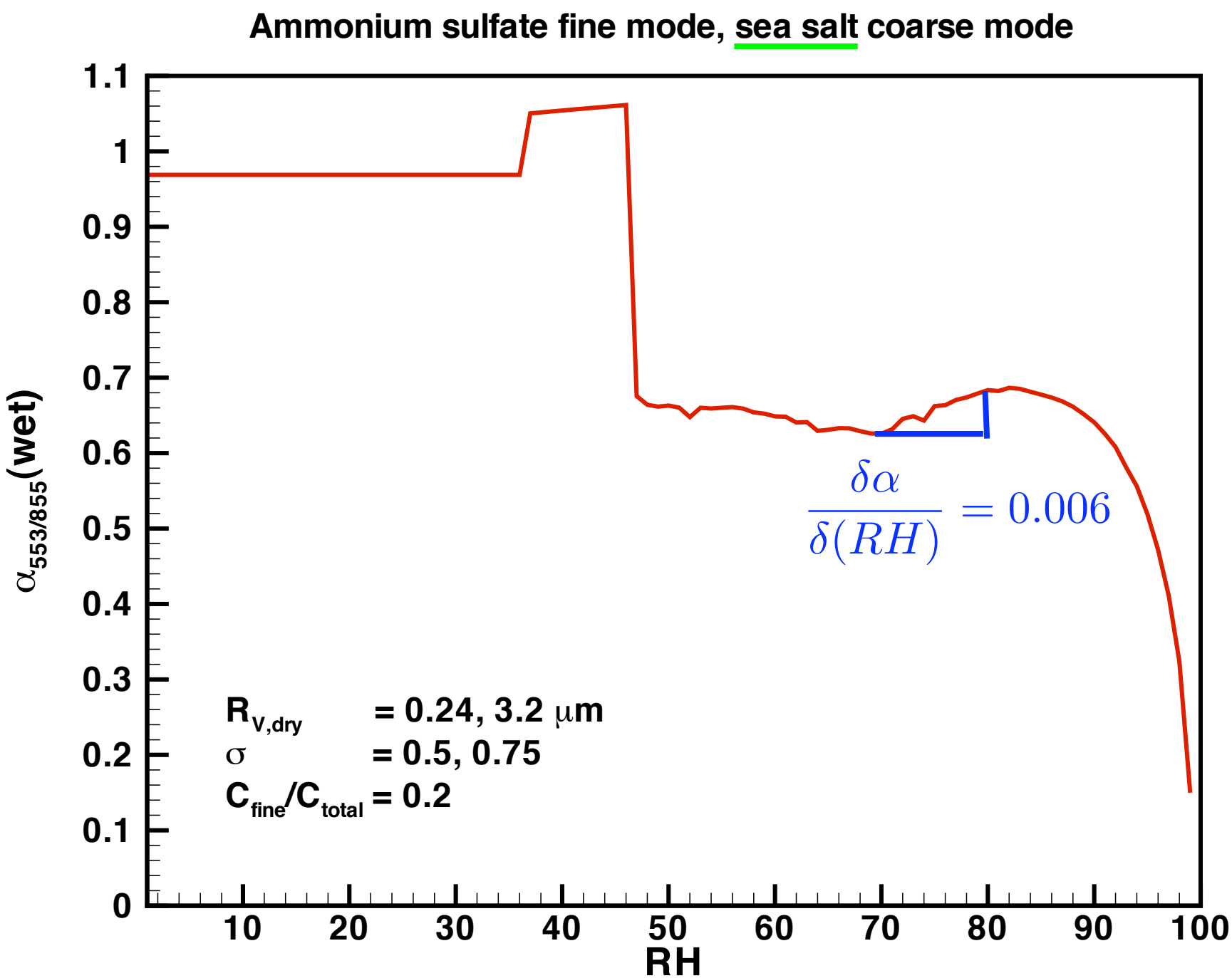

Remote Continental Aerosols
RH = 98% for wet aerosols
0.8
0.6
0.4
0.2
0
80
60
40
20
0
dV_dlnr_nuclei
dV_dlnr
wet accum mode
coarse mode
wet nuclei mode
dry accum mode
dry nuclei mode
10^{-2}
10^{-1}
10^{0}
10^{1}
radius


OPAC Water soluble and sea salt accumulation
(No RH growth in coarse mode)
trimode
bimode
radius accum = 0.058
radius coarse = 0.320
log sigma accum = 0.217
log sigma coarse = 0.380
AE_646-855(wet)
RH
0
0.2
0.4
0.6
0.8
1
1.2
20
40
60
80
100


Ammonium sulfate accumulation mode, sea salt coarse mode
dry
wet fit, rh = 46
wet fit, rh = 47
$R_{V, dry}$ = 0.24, 3.2 μm
σ = 0.5, 0.75
C_{fine}/C_{total} = 0.2
dV/dlnr
Radius μm
2
1
0
10^{-3}
10^{-2}
10^{-1}
10^{0}
10^{1}
10^{2}

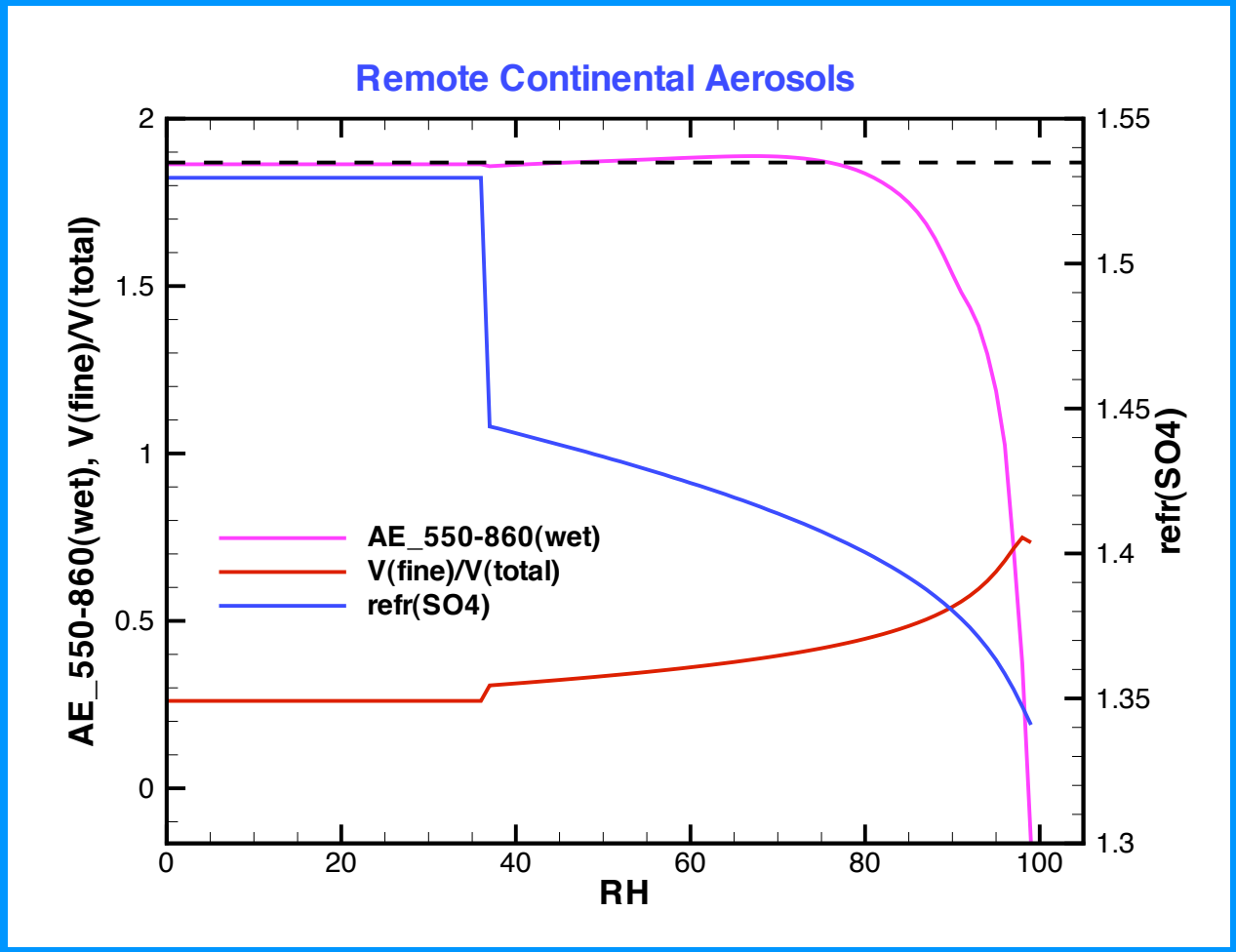

Ammonium sulfate accumulation mode, sea salt coarse mode
2
1.5
1
0.5
0
-0.5
-1
-1.5
-0.5
-0.25
0
$\alpha_{99} - \alpha_{80}$
$\alpha_{80} - \alpha_{40}$
V_{fine} / V_{total}
at RH=40
0.9
0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1


Ammonium sulfate accumulation mode, dust coarse mode
volum_finefrctn_rh40
0.9
0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1
1
0
0
1
2
3
4
$\alpha_{80} - \alpha_{40}$
α_{40}


Ammonium sulfate fine mode, sea salt coarse mode
$\alpha_{553/855}$(wet)
RH
$\frac{\delta\alpha}{\delta(RH)} = 0.006$
$R_{V,dry}$ = 0.24, 3.2 μm
σ = 0.5, 0.75
C_{fine}/C_{total} = 0.2

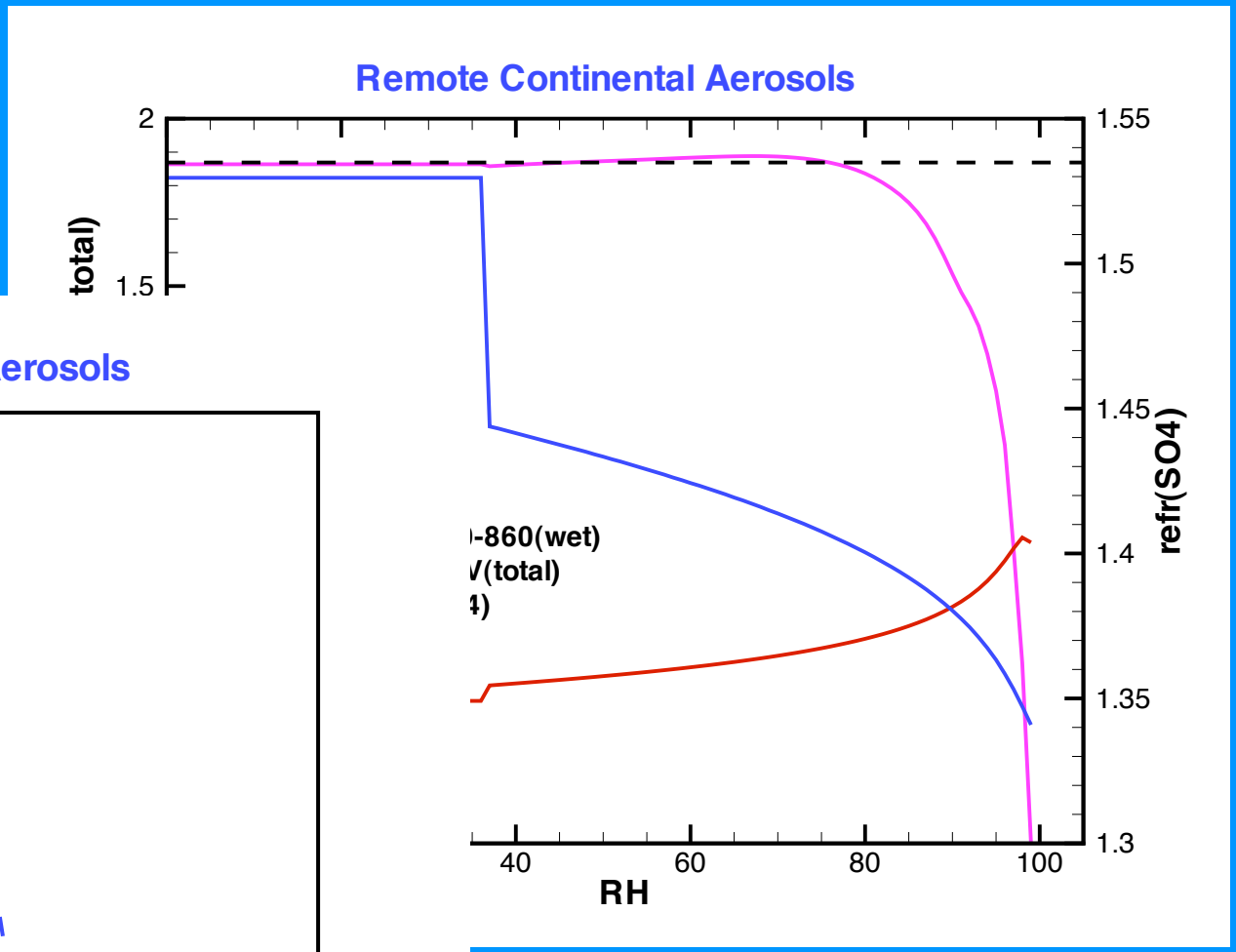
Impact of relative humidity on a Jaenicke (1993) size distribution

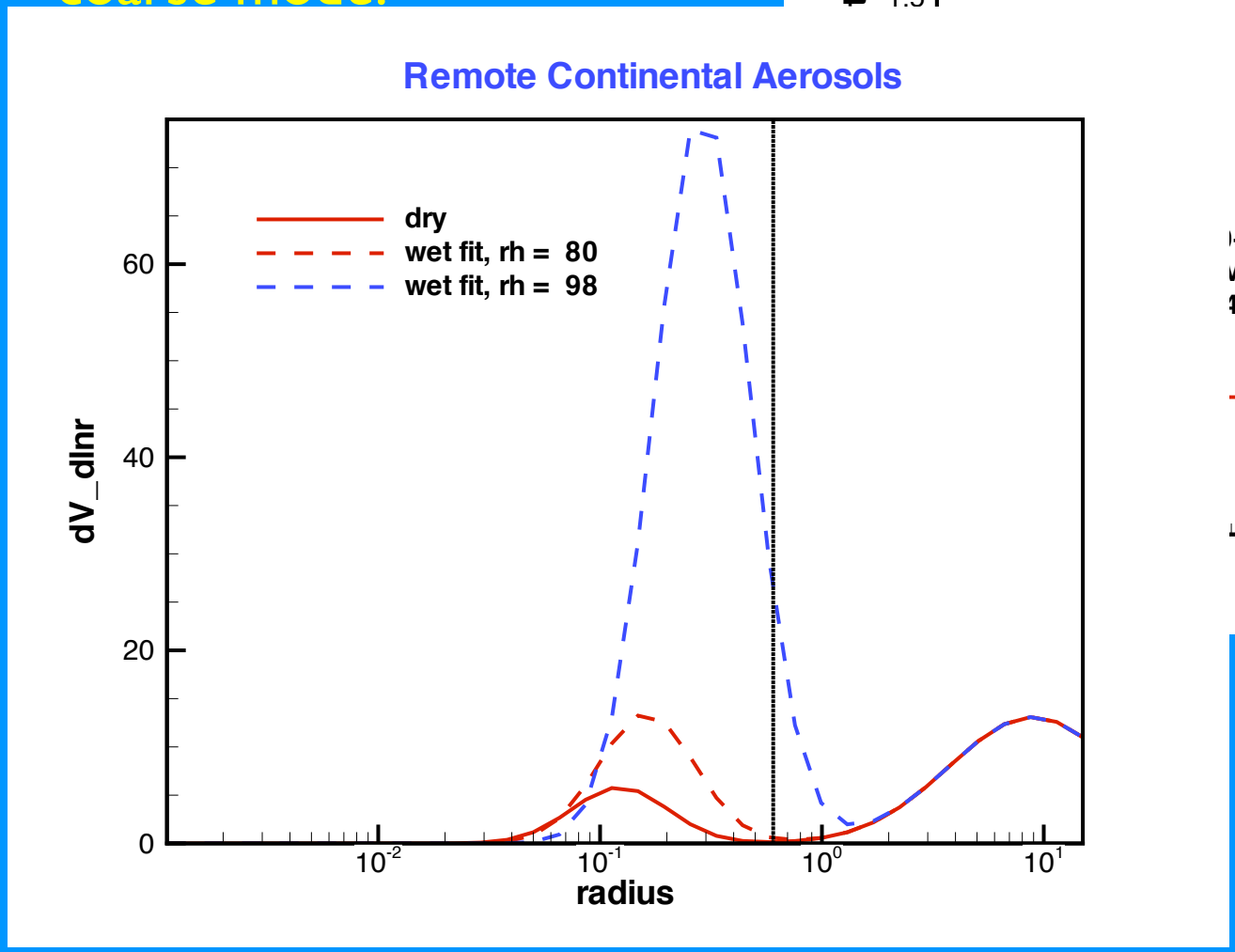
- Fine mode allowed to grow with RH.
- No growth allowed in coarse mode.
- Dry refractive index = 1.53 10^{-7}.

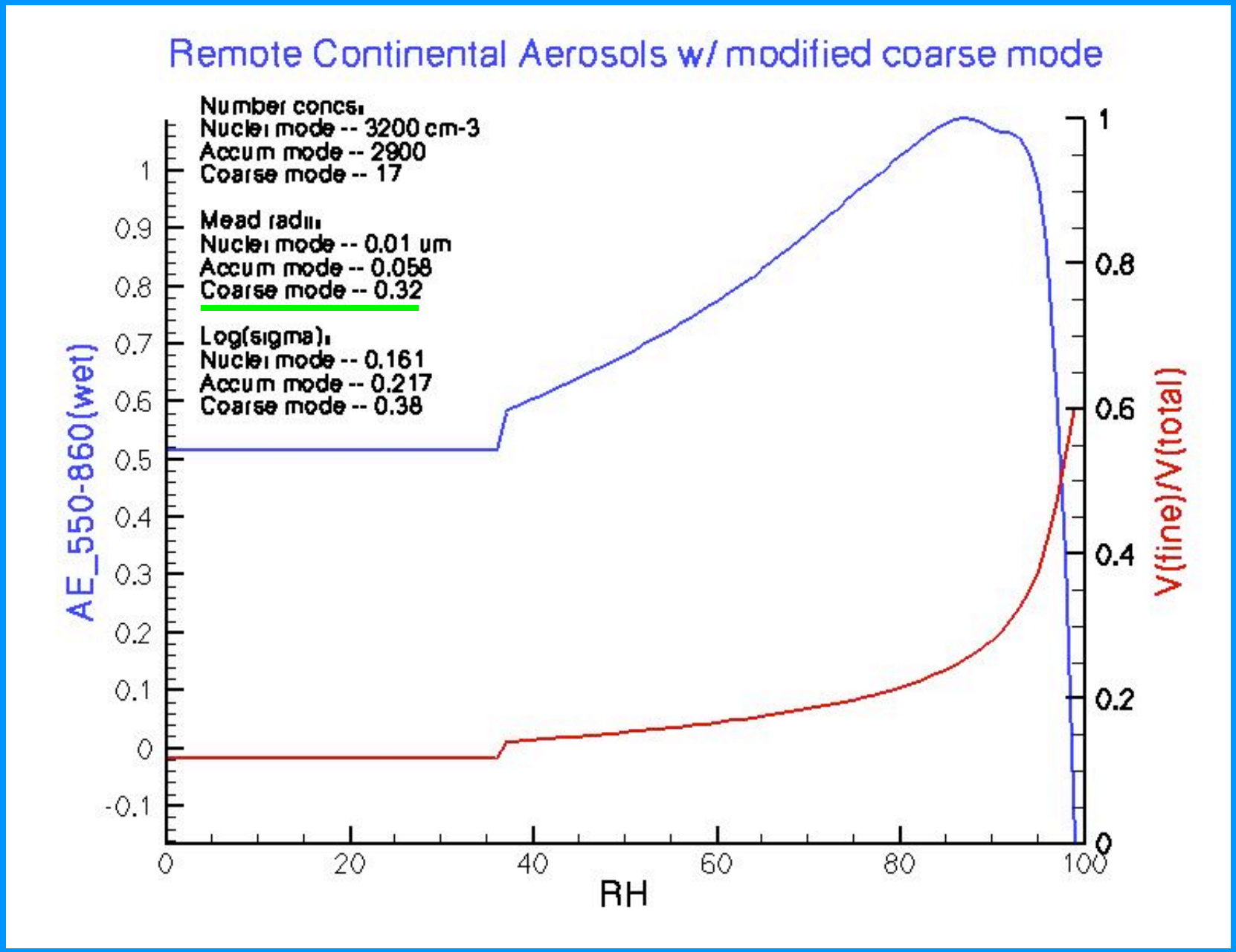


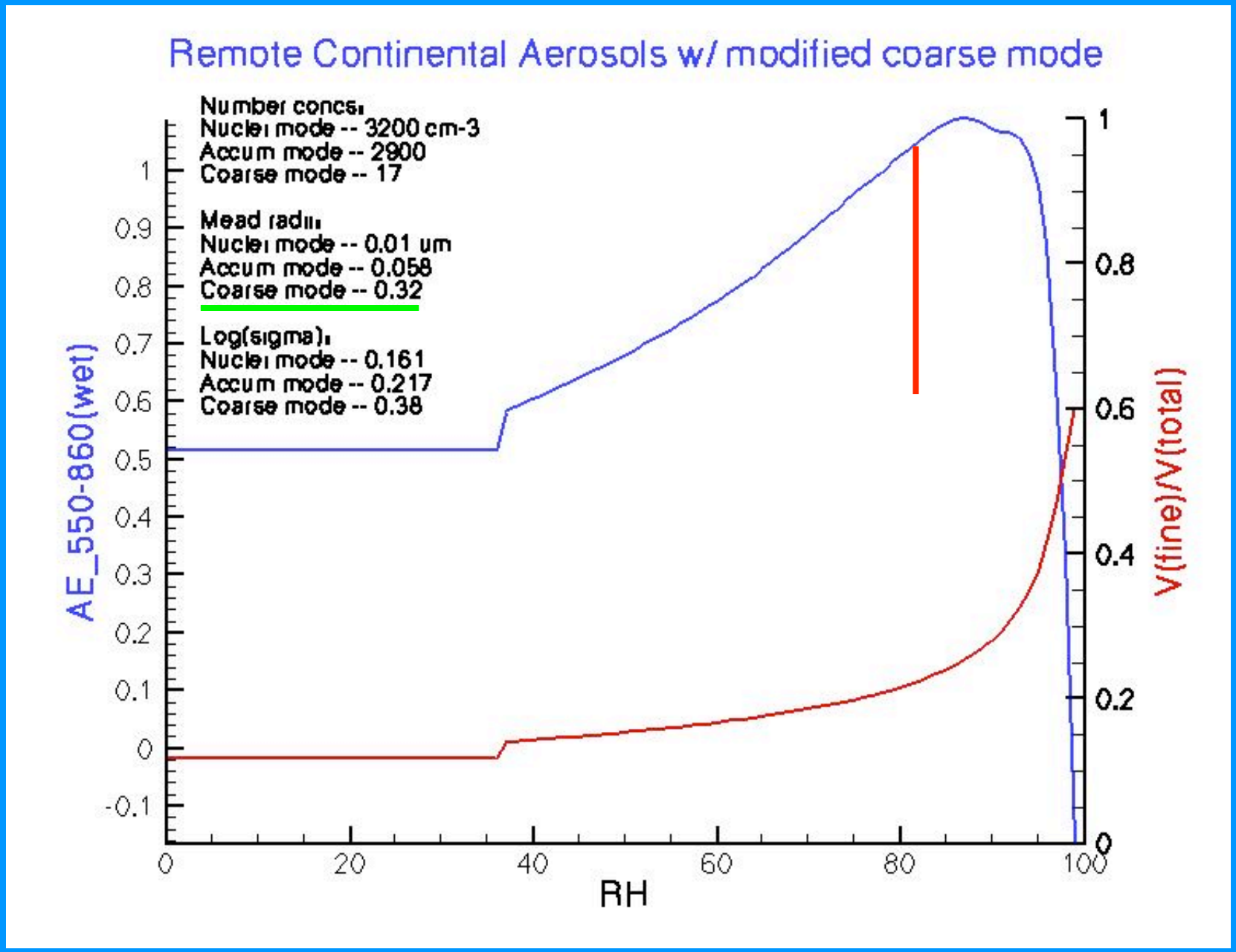
Impact of relative humidity on a Jaenicke (1993) size distribution

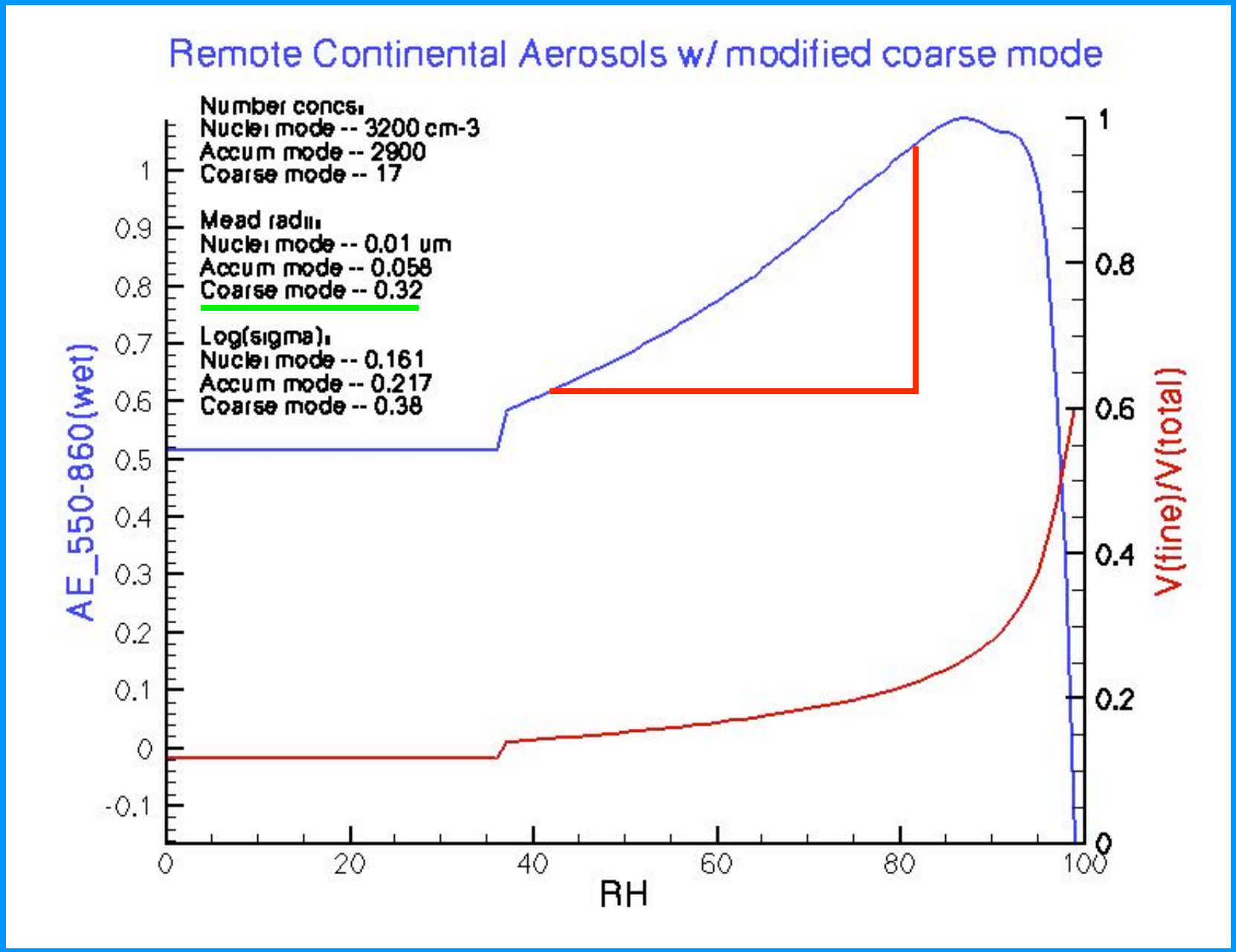
- Fine mode allowed to grow with RH.
- No growth allowed in coarse mode.

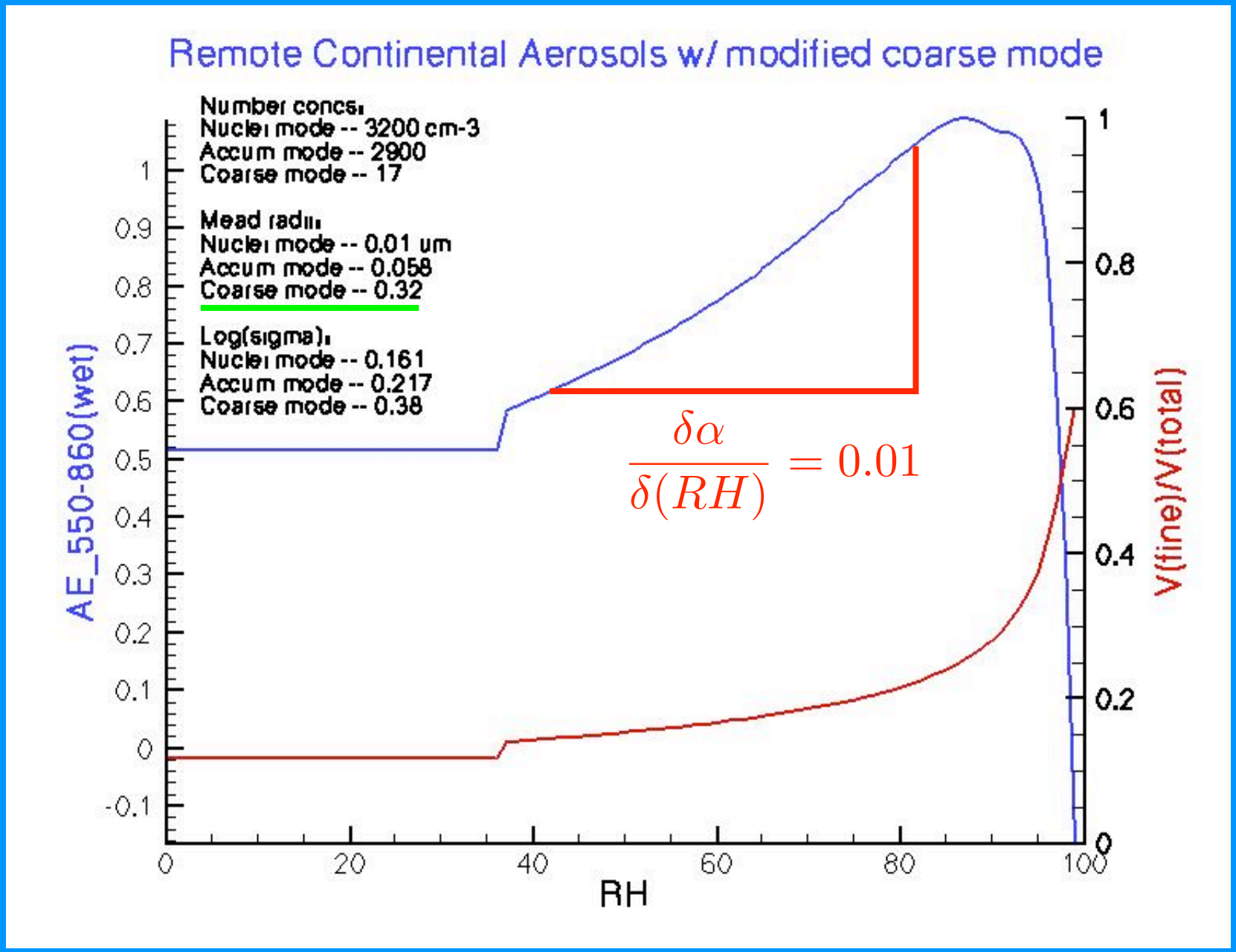


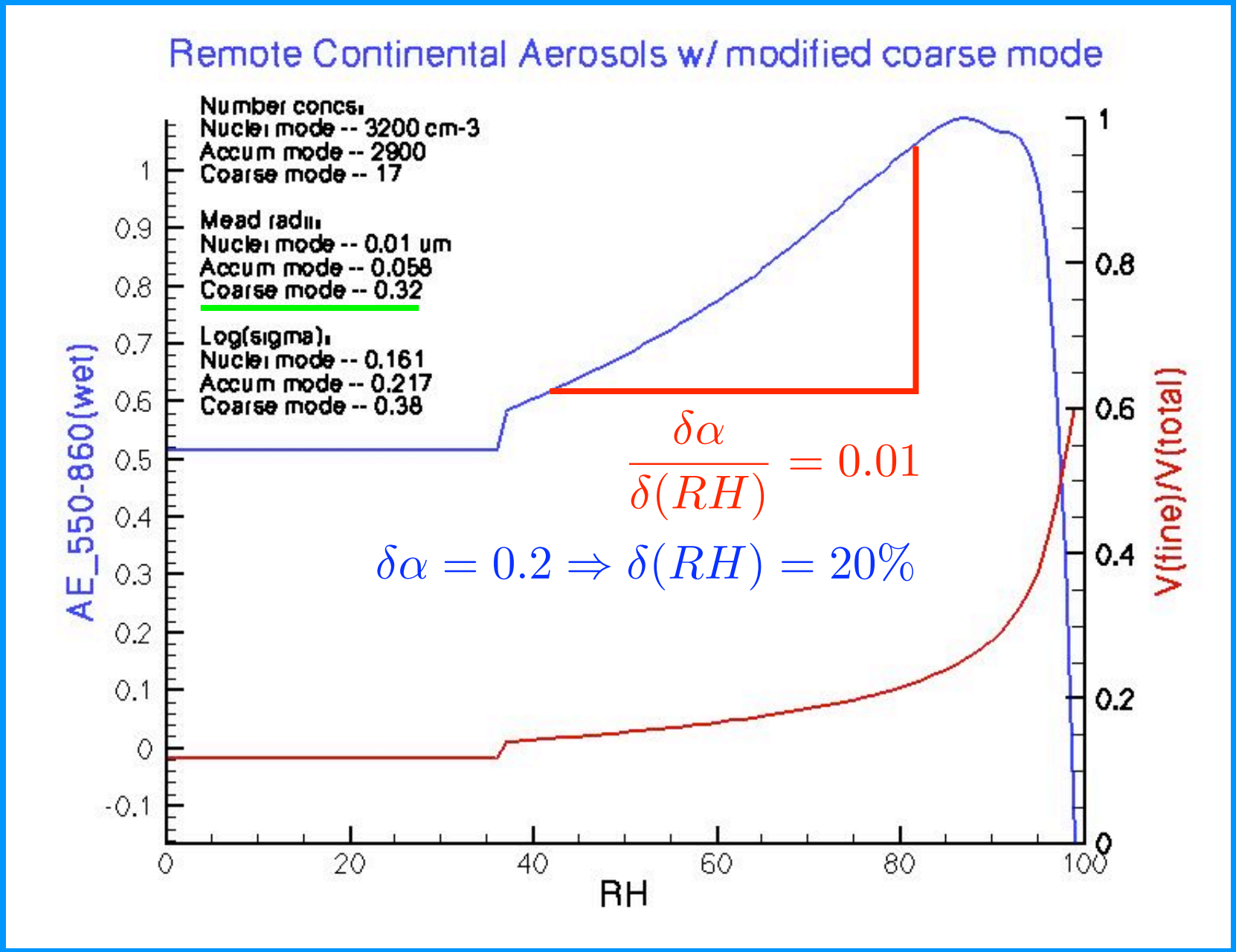



Remote Continental Aerosols w/ modified coarse mode
Number concs:
Nuclei mode -- 3200 cm-3
Accum mode -- 2900
Coarse mode -- 17
Mead radii:
Nuclei mode -- 0.01 um
Accum mode -- 0.058
Coarse mode -- 0.32
Log(sigma):
Nuclei mode -- 0.161
Accum mode -- 0.217
Coarse mode -- 0.38
AE_550-860(wet)
V(fine)/V(total)
RH
1
0.9
0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1
0
-0.1
0
20
40
60
80
100
1
0.8
0.6
0.4
0.2
0

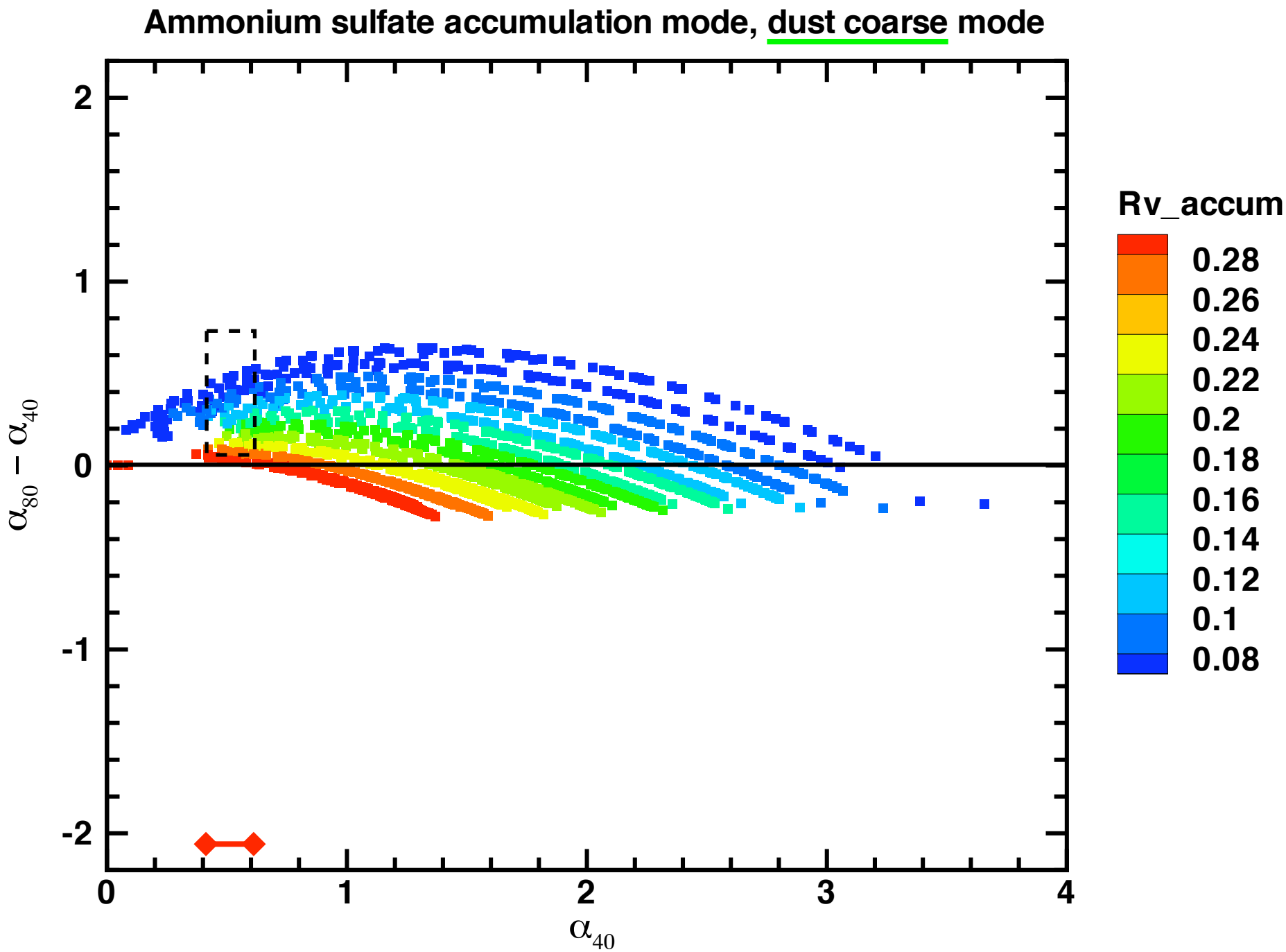

Remote Continental Aerosols w/ modified coarse mode
Number concs:
Nuclei mode -- 3200 cm-3
Accum mode -- 2900
Coarse mode -- 17
Mead radii:
Nuclei mode -- 0.01 um
Accum mode -- 0.058
Coarse mode -- 0.32
Log(sigma):
Nuclei mode -- 0.161
Accum mode -- 0.217
Coarse mode -- 0.38
AE_550-860(wet)
V(fine)/V(total)
RH
1
0.9
0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1
0
-0.1
0
20
40
60
80
100
1
0.8
0.6
0.4
0.2
0

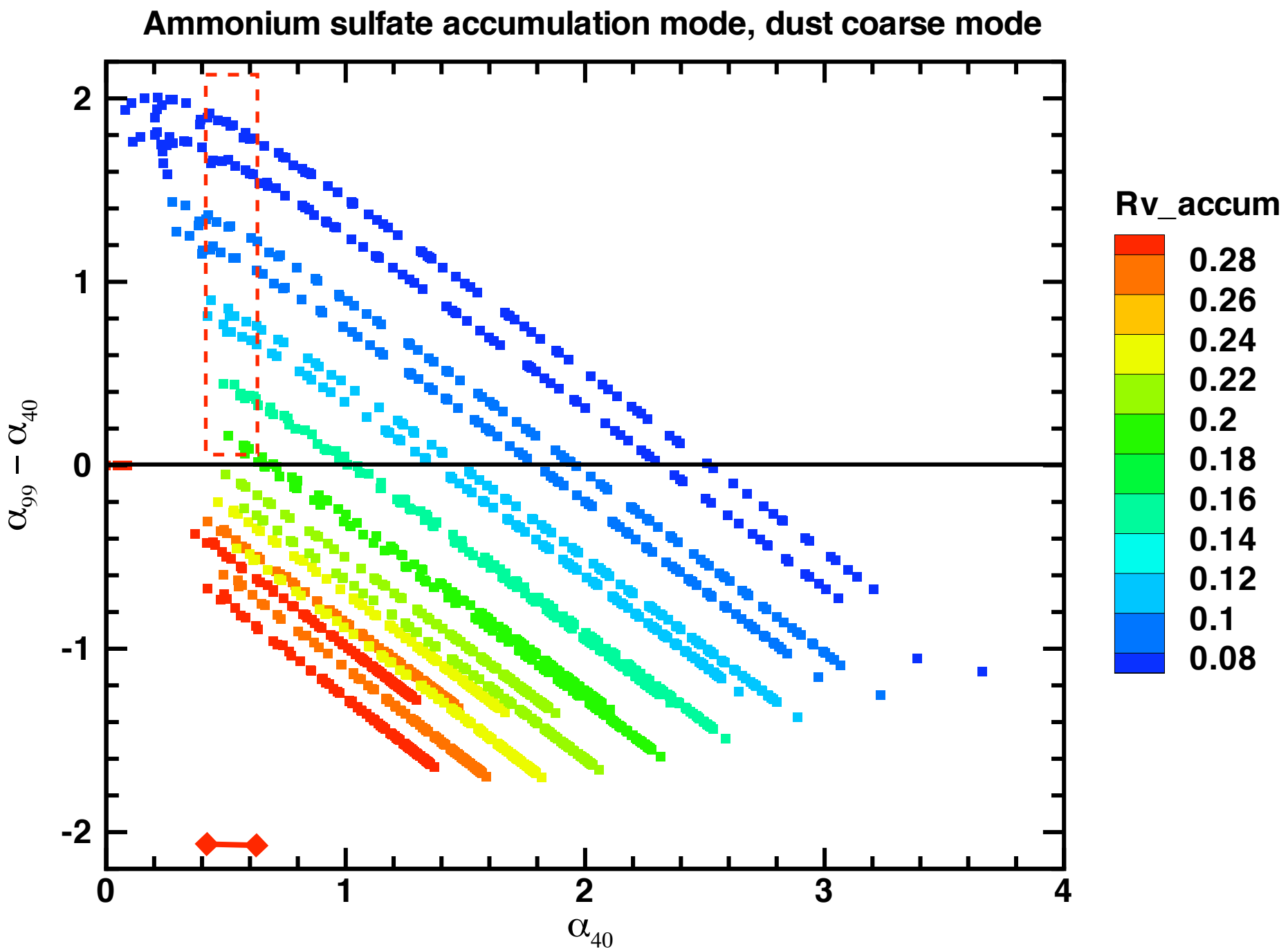

Remote Continental Aerosols w/ modified coarse mode
Number concs:
Nuclei mode -- 3200 cm-3
Accum mode -- 2900
Coarse mode -- 17
Mead radii:
Nuclei mode -- 0.01 um
Accum mode -- 0.058
Coarse mode -- 0.32
Log(sigma):
Nuclei mode -- 0.161
Accum mode -- 0.217
Coarse mode -- 0.38
AE_550-860(wet)
V(fine)/V(total)
RH
1
0.9
0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1
0
-0.1
0
20
40
60
80
100
1
0.8
0.6
0.4
0.2
0

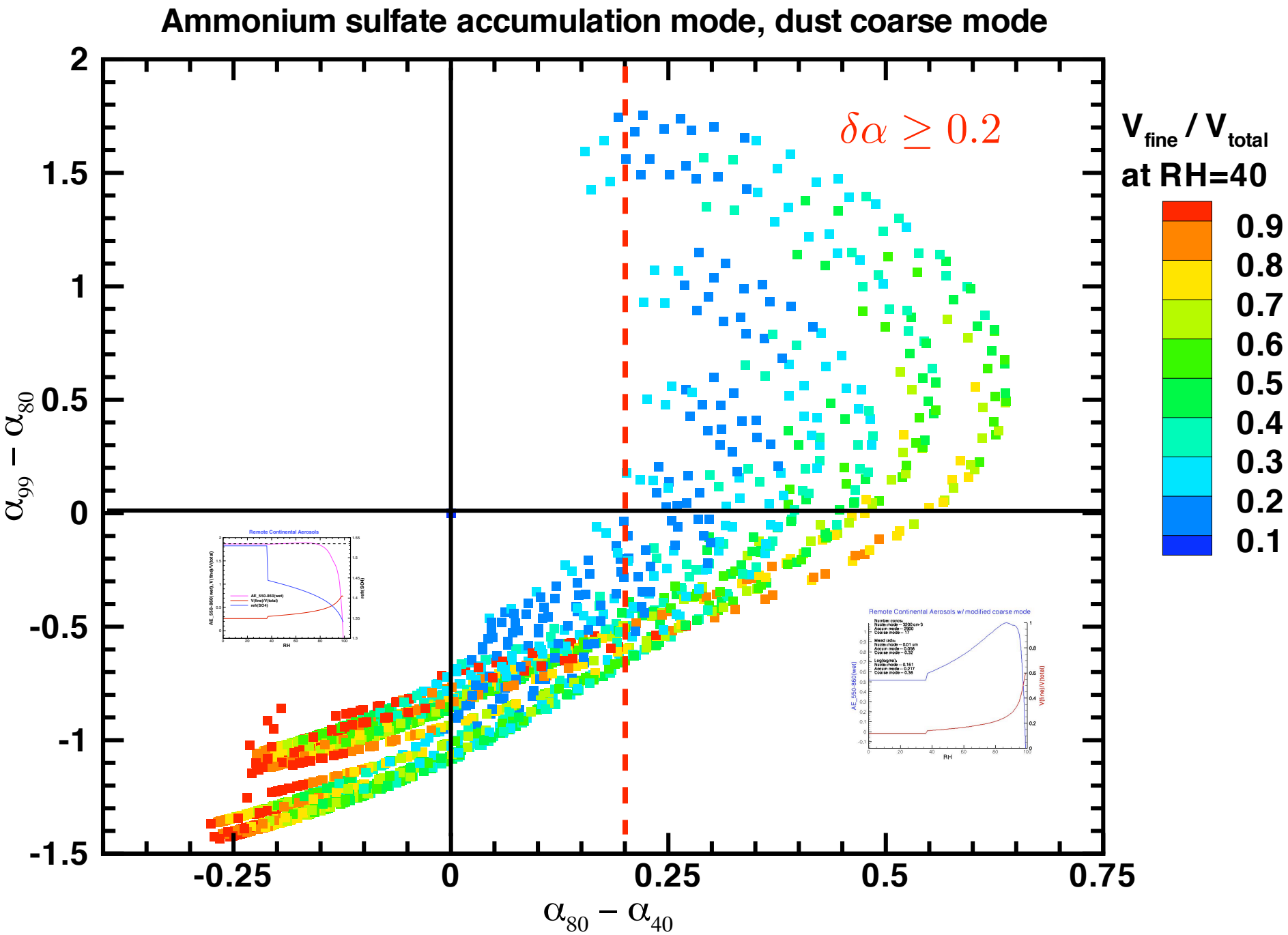

Remote Continental Aerosols w/ modified coarse mode
Number concs:
Nuclei mode -- 3200 cm-3
Accum mode -- 2900
Coarse mode -- 17
Mead radii:
Nuclei mode -- 0.01 um
Accum mode -- 0.058
Coarse mode -- 0.32
Log(sigma):
Nuclei mode -- 0.161
Accum mode -- 0.217
Coarse mode -- 0.38
$\frac{\delta\alpha}{\delta(RH)} = 0.01$
AE_550-860(wet)
V(fine)/V(total)
RH
1
0.9
0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1
0
-0.1
0
20
40
60
80
100
1
0.8
0.6
0.4
0.2
0

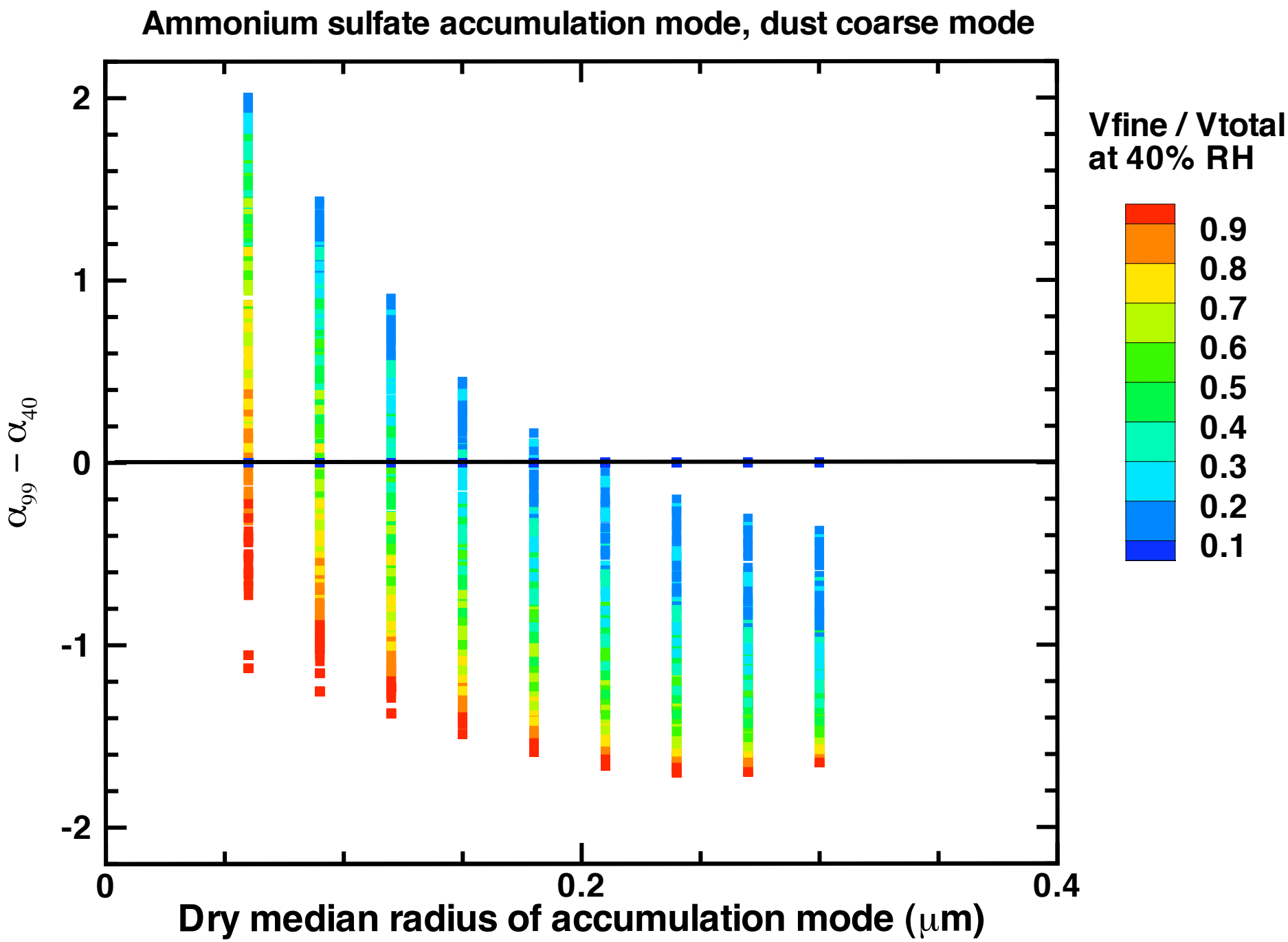

Remote Continental Aerosols w/ modified coarse mode
Number concs:
Nuclei mode -- 3200 cm-3
Accum mode -- 2900
Coarse mode -- 17
Mead radii:
Nuclei mode -- 0.01 um
Accum mode -- 0.058
Coarse mode -- 0.32
Log(sigma):
Nuclei mode -- 0.161
Accum mode -- 0.217
Coarse mode -- 0.38
AE_550-860(wet)
V(fine)/V(total)
RH
$\frac{\delta\alpha}{\delta(RH)} = 0.01$
$\delta\alpha = 0.2 \Rightarrow \delta(RH) = 20\%$

Part I
Ammonium Sulfate Fine Mode,
Dust Coarse Mode


Ammonium sulfate accumulation mode, dust coarse mode
2
1
0
-1
-2
0
1
2
3
4
$\alpha_{80} - \alpha_{40}$
α_{40}
Rv_accum
0.28
0.26
0.24
0.22
0.2
0.18
0.16
0.14
0.12
0.1
0.08


Ammonium sulfate accumulation mode, dust coarse mode
Rv_accum
0.28
0.26
0.24
0.22
0.2
0.18
0.16
0.14
0.12
0.1
0.08
2
1
0
-1
-2
0
1
2
3
4
$\alpha_{99} - \alpha_{40}$
α_{40}


Ammonium sulfate accumulation mode, dust coarse mode
$\delta\alpha \geq 0.2$
V_{fine} / V_{total}
at RH=40
0.9
0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1
$\alpha_{99} - \alpha_{80}$
$\alpha_{80} - \alpha_{40}$
2
1.5
1
0.5
0
-0.5
-1
-1.5
-0.25
0
0.25
0.5
0.75
Remote Continental Aerosols
Remote Continental Aerosols w/ modified coarse mode
RH


Ammonium sulfate accumulation mode, dust coarse mode
2
1
0
-1
-2
$\alpha_{99} - \alpha_{40}$
0
0.2
0.4
Dry median radius of accumulation mode (μm)
Vfine / Vtotal
at 40% RH
0.9
0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1