Modeling aerosol optical depth: What are the limitations of a bulk aerosol scheme?

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Representation of aerosol size in GEOS-Chem

- Bulk scheme carries mass only, assumes size distribution offline for optics.
- Sectional scheme (TOMAS) allows mass transfer between 40 bins through coagulation. Internally mixed.
- AOD ($\tau$) is dependent on the surface area and the Mie extinction efficiency.

$$\tau \propto Q_{\text{ext}}(r)\pi r^2$$
- Harmonized emissions used for bulk and sectional schemes.
- TOMAS AOD 25% lower globally
- Lower dust and sea-salt AOD and higher AOD from BC and OC
Column (total) mass burden

- Sectional mass burden ~8% higher than Bulk
- ~10% more dust, sea-salt and SO4 in sectional scheme
- ~10% less OC and BC in sectional scheme
How do aerosol size differences influence the AOD?

- TOMAS OC at larger sizes contributes to higher AOD
- Less dust mass at optically active smaller size in TOMAS giving lower AOD
- Less sea-salt (and associated water) concentrated at smaller sizes in TOMAS gives higher AOD
Sea-salt emissions and size distribution

- TOMAS distribution follows the emissions
- Bulk distribution forced to smaller size
- High surface area of bulk sea-salt yields double AOD relative to TOMAS AOD
Aerosol Water

- Water contributes between 28% and 79% of AOD based on AEROCOM models.
- Water contributes 47% in bulk and 52% in TOMAS.

**BULK**

- AOD = 0.81

**SECTIONAL**

- AOD = 0.61

Fractional increase in TOMAS AOD from water:

- SO4
- Sea-Salt
- BC
- OC
- Dust
- H2O
Effect of upper limit on RH on the AOD

- Importance of water is magnified further depending on the upper limit of RH...
- 99% cap gives 40% higher global AOD than 95% cap
- >90% of (hourly) cases with RH>99% are cloud-free i.e. a real effect on AOD
Conclusions

• Regional monthly AOD can vary by -40% / +100% both at source and downwind between bulk and sectional schemes with matching emissions
  • Differences driven more by size differences than mass differences between schemes

• Discrepancy between emitted size and assumed size for optics for sea-salt and dust
  • Sea-salt coarse mode size needs updating
  • Dust online bulk submicron bin size need altering
  • Comparison with observed size distributions will determine if TOMAS correct

• Aerosol water generally well-constrained, but strongly influences AOD
  • RH upper limit choices lead to large AOD difference (may explain large diversity in multi-model studies)
Dust emissions and size distribution

• TOMAS distribution follows the emissions

• Bulk distribution forced to smaller size

• High surface area of bulk dust yields 40% higher AOD relative to TOMAS AOD

Bulk scheme places more dust mass at submicron sizes than TOMAS suggests
Mass extinction efficiency (MEE)

- Similar global average MEE between bulk and sectional (1.45 vs 1.31 m²/g).
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• Strong differences for individual species

• Spatial structure indicates size-dependent effect on AOD
GADS water uptake factors

Water Uptake – Hygroscopic growth uncertainties

- Relatively small difference from different water uptake methods
- Water uptake parameterization yields small (~5%) bias on the global AOD
- 87% of the difference still remains for BULK and TOMAS with identical water uptake
- Locally, absolute differences range from -0.09 (-33%) to +0.07 (+100%) between GADS, TOMAS and ISOROPPIA

Aerosol water exacerbates differences, but not the main cause
• Importance of water is magnified further depending on the upper limit of RH...
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Humidity cap pretty important!