Precipitation Scavenging: Search for a Physically-Based Algorithm in the UCI CTM

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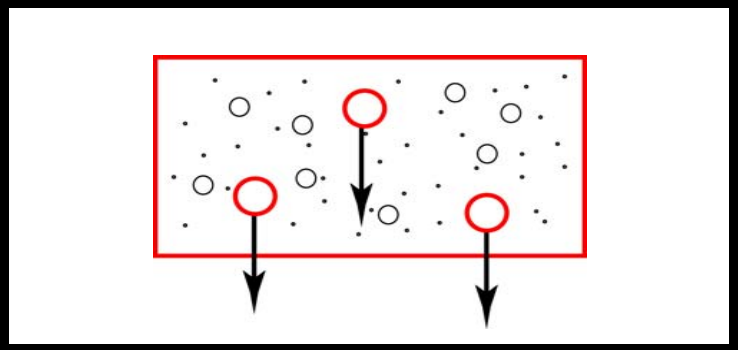
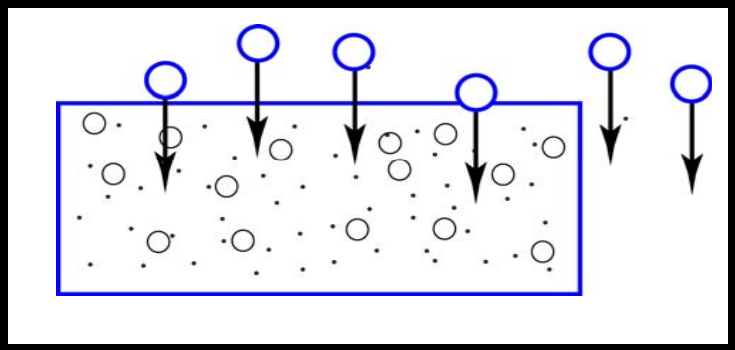
In-Cloud Scavenging (rainout, nucleation scavenging)



Local uptake by initial cloud droplets and their conversion to precipitation

Scavenging proportional to amount of condensate converted to precipitation

Below-Cloud Scavenging (washout, impaction scavenging)



Collection by falling droplets, either from interstitial / ambient air (most common) or liquid via accretion processes (e.g. Rotstayn, 1997)

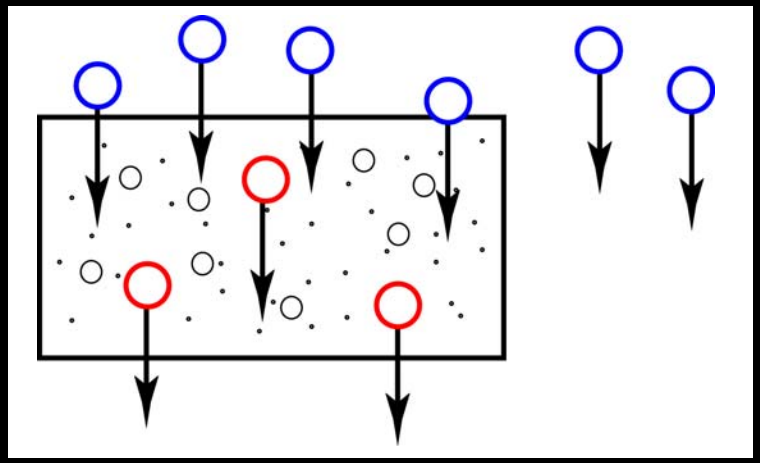
Scavenging proportional to precipitation flux in the layer

Both modeled as a first-order loss process: $X_{iscav} = X_i F\,(1 - \exp(-\lambda \Delta t))$

Loss rate depends on cloud water, rate of precipitation formation, and rate of tracer uptake by liquid phase

Loss rate depends on precipitation rate and rate of tracer uptake by the liquid phase, mass-transfer rate, or collision rate, depending on species

$$X_{iscav} = X_i F\,(1 - \exp(-\lambda \Delta t))$$



$F_{InCloud}$

Tracer is removed from the fraction of the cloud that is converted to precipitation

$$F_{max} = CF$$

Giorgi and Chameides (1986):

$$F = (CF)\,Q\Delta t/(CW)\,kT_C$$

Q: Precip formation rate k: rate of conversion of CW to precip
Tc: duration of precip over timestep

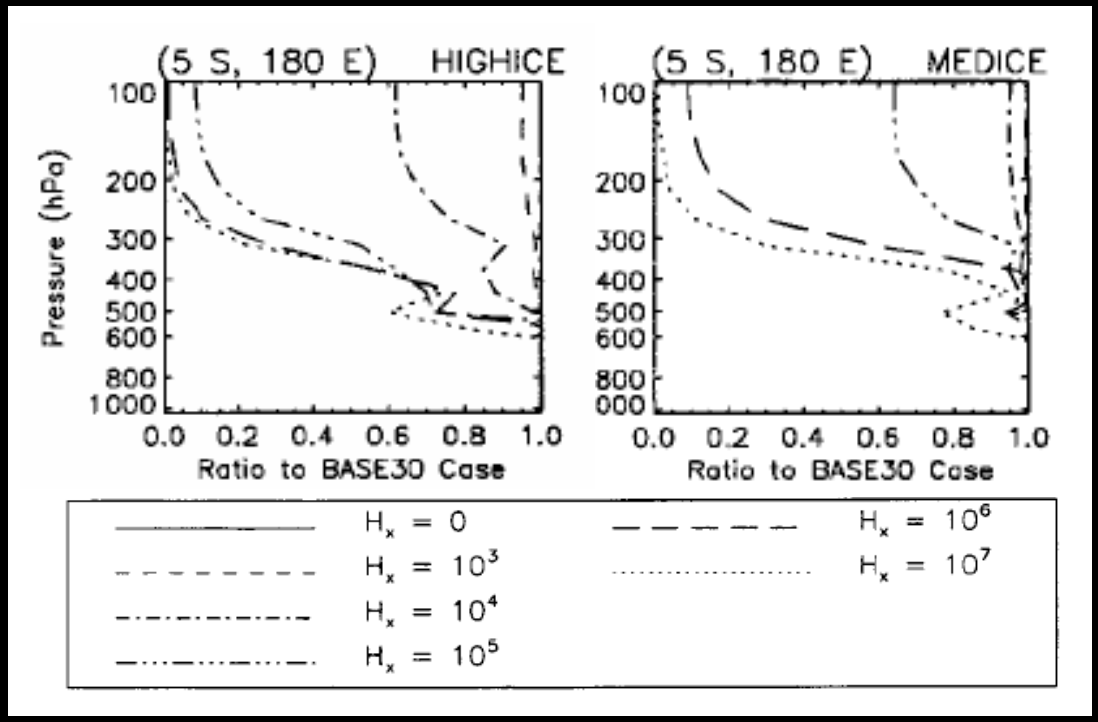
$F_{BelowCloud}$

More difficult to determine - Some models use the largest overhead cloud fraction (GEOS-CHEM, MATCH), some use fixed F, some assume that all of the rain falls within the local cloud fraction, independent of cloud above

$$F_{max} = 1.0$$

	Transfer into Condensed Phase	In-Cloud Scavenging	Below-Cloud Scavenging
Moderately Soluble Gases	Liquid uptake limited by Henry's Law - depends on pH and T. Probably no uptake by ice (e.g. Diehl, 1998)	Limited by both Henry's Law and precip formation rate	Limited by both Henry's Law and precip rate
Highly Soluble Gases	Liquid uptake theoretically limited by Henry's Law (+dissociation), but generally complete. Uptake by ice likely depends on dissociation (HONO2 vs H2O2)	In effect limited by precip formation rate	Kinematically limited (mass-transfer)
Aerosols	Fully dissolved in liquid phase. Ice??	Limited by precip formation rate	Kinematically limited (collision)

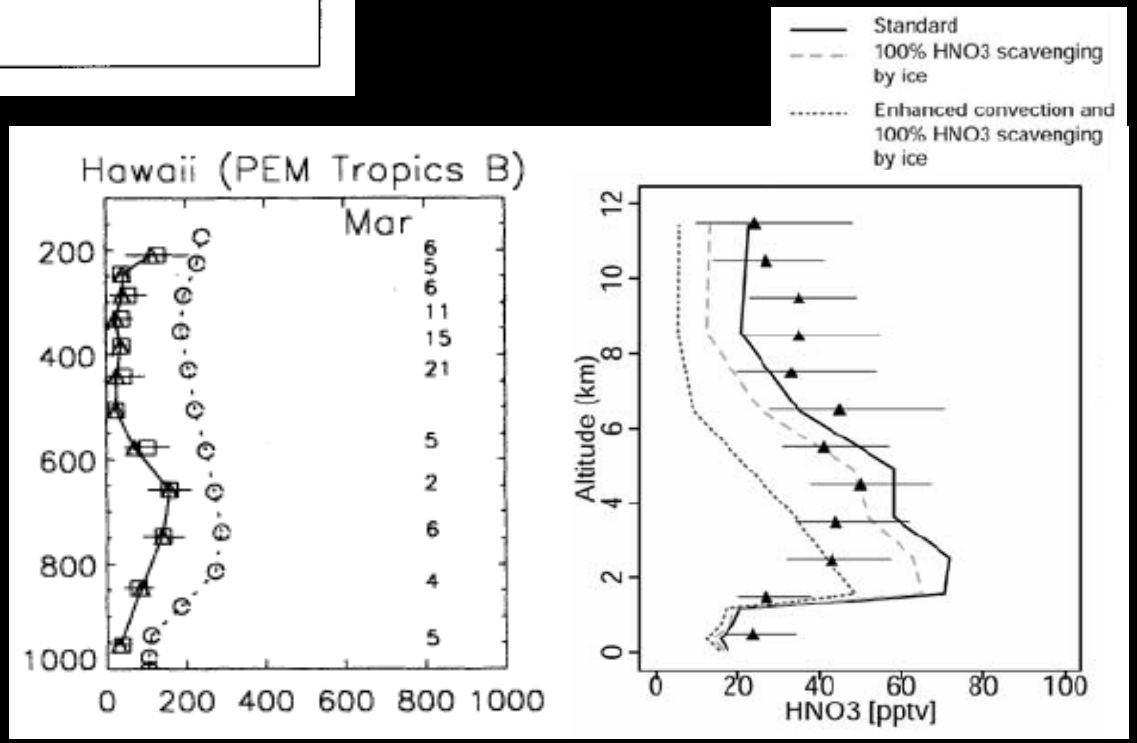
	In-Cloud Scavenging	Below-Cloud Scavenging
Liquid Precipitation	Initial droplet growth by collision – coalescence. Fraction of tracer in the liquid phase is equally distributed among cloud water and fully incorporated as rain drops form	Usually treated as rain falling through interstitial or ambient air and incorporating tracer from vapor phase. Rotstayn and Lohmann (2002) parameterize droplet collision and growth.
Frozen Precipitation	Initial growth either treated the same as liquid precip (too efficient) or ignored (no removal) – neither seems to be correct. Some models include cirrus gravitaitonal settling – uniform, all ice particles (e.g. Crutzen and Lawrence, 2000)	Generally treated the same as liquid precip or ignored. Rotstayn and Lohmann (2002) parameterize accretion by snow. For a given precip flux, snow scavenges a larger area than rain (smaller D_{eff}), but has a smaller collection efficiency



Crutzen and Lawrence (2000):
Ratio of idealized tracer from runs with
$SCAV_{ice} = SCAV_{Liquid}$
and
$SCAV_{ice} = 0.1 * SCAV_{Liquid}$
to run with
$SCAV_{ice} = 0$

Bey et al. (2001): HONO2 from GEOS-CHEM with no in-cloud ice scavenging, compared to PEM-Tropics B observations

Staudt et al. (2003): Sensitivity of GEOS-CHEM to efficiency of ice scavenging in updrafts



Given CF(L) and R(L), how do we partition precipitation into in-cloud and below-cloud scavenging?

This matters because rainout is much more efficient than washout

How can we treat ice scavenging in a physical way that removes tracers at a rate consistent with observations?

We assume that precipitating clouds are maximally overlapped, and that if there is precipitation in a layer, at least 20% of that layer has condensed water

We maintain a precipitation “core” that contains aged precipitation (min 20%)

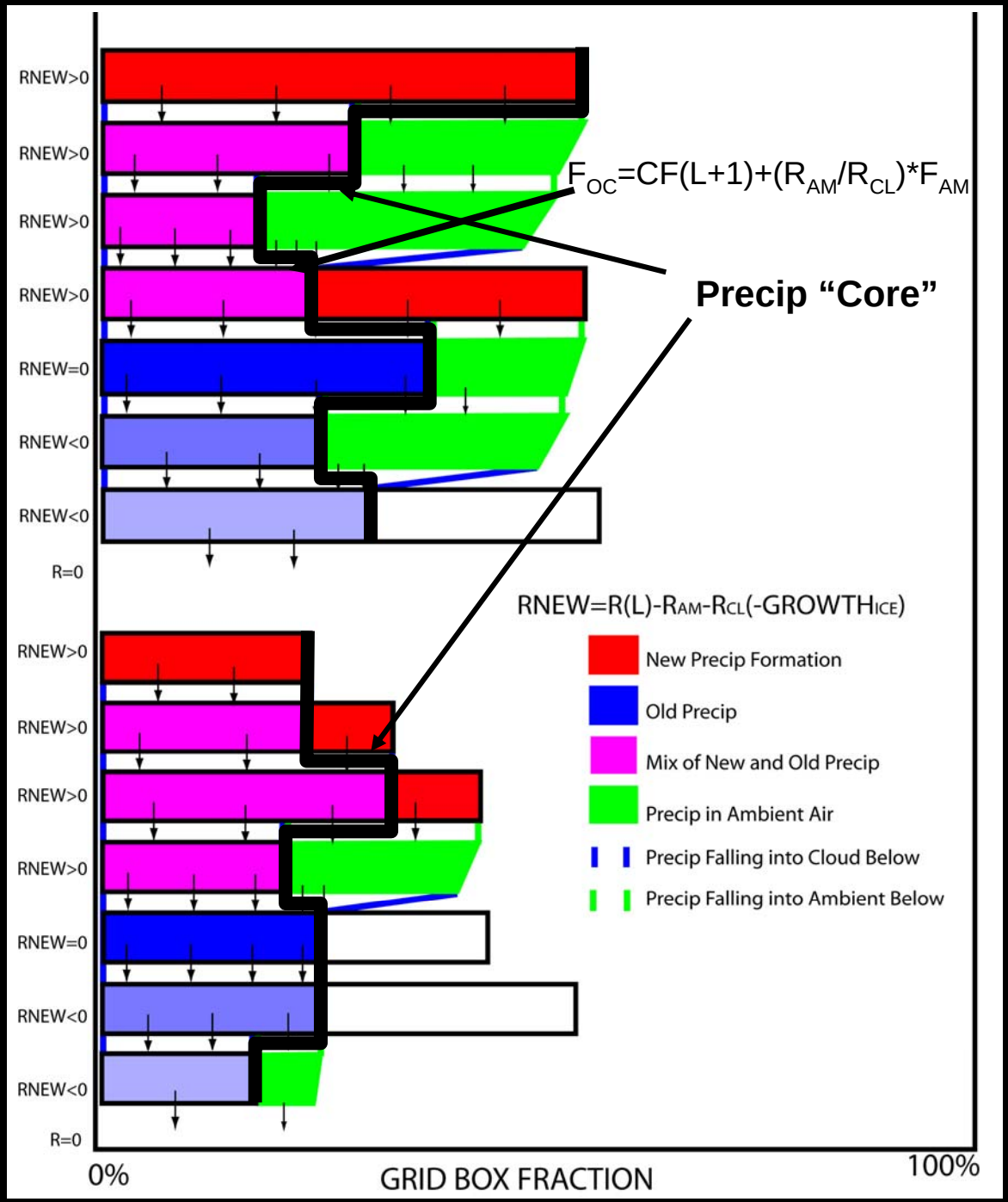
Each model level is partitioned into up to 4 sections, each with a gridbox fraction, precipitation rate, and precipitation diameter:

Old Cloud – Area of the gridbox with cloud that also has rain falling from above Cloud “core” – aged precipitation	New Cloud – Area of the gridbox with cloud and no rain falling from above	Ambient – Area of the gridbox with rain from above falling through clear sky Constant rate of evaporation – reduces both area and rain amount	Clear Sky – Area of the gridbox with no cloud and no rain from above

New precip is spread evenly between OC and NC

For the next level, we combine the 4 sections and line them up with the cloudy and clear regions below to generate 4 new sections

Similar to Jakob and Klein (2000)



$$X_{iscav} = X_i F\,(1 - \exp(-\lambda \Delta t))$$

New Rain:

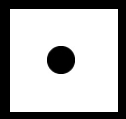
$$F = CF\,(1 - \exp(-(R_{NEW}/CW)\Delta t))$$

New Ice:

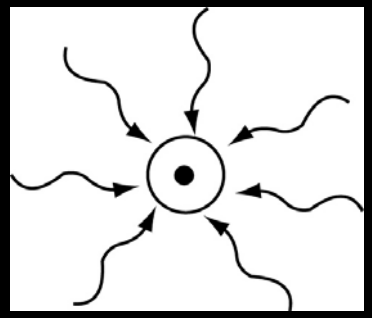
$$F = CF$$

Old Rain / Ice:

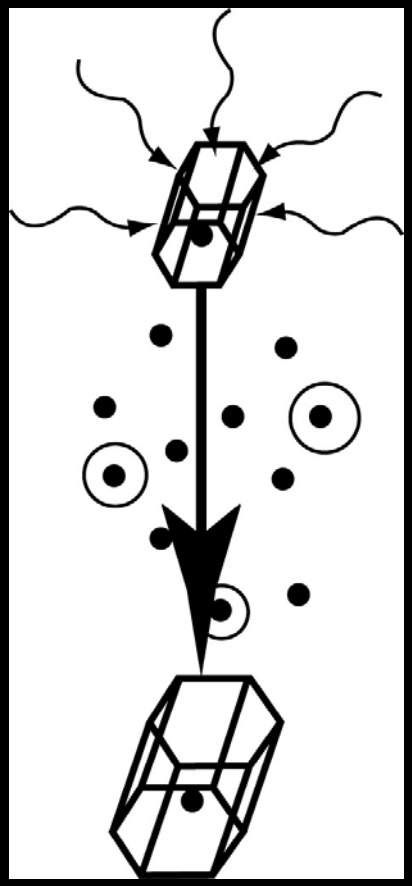
$$F = F_{OC} \text{ or } F_{AM}$$



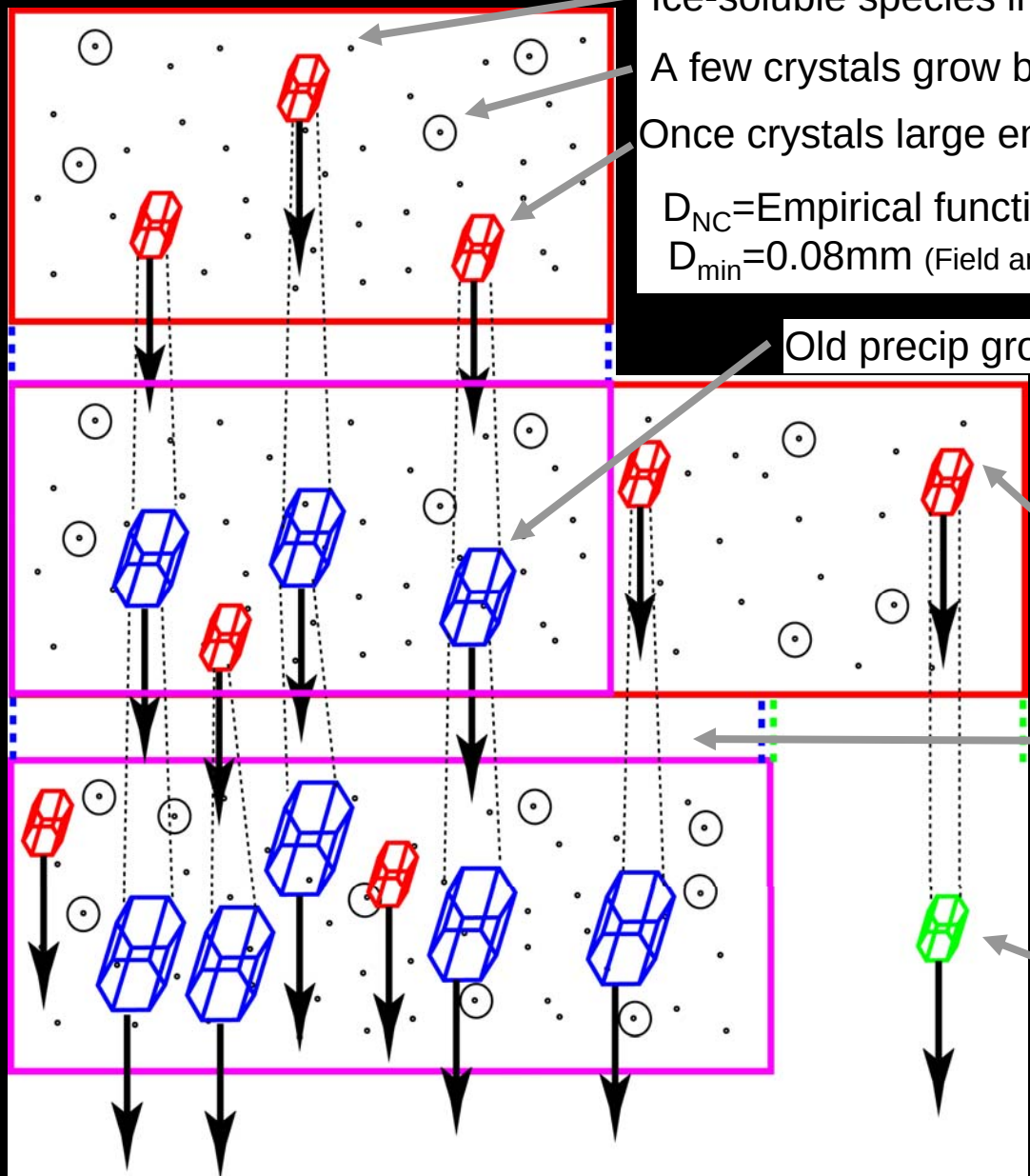
1. Ice nucleus is formed by nucleation from the vapor phase or drop freezing



2. Small ice particles grow primarily via vapor deposition (Alheit et al., 1990; Field and Heymsfield, 2002)



3. Larger ice particles (>100 micron) grow primarily by aggregation and riming. Riming dominates scavenging (Alheit et al., 1990) Only particles in this size range can irreversibly remove HNO3 (Tabazedeh et al., 1999), and it is removed primarily on the surface of the ice – bulk uptake is negligible (Sommerfeld et al., 1998)



Ice-soluble species in condensed phase (Ice Nuclei)

A few crystals grow by vapor deposition – no removal

Once crystals large enough to fall (R(L)>0) – impact scavenging

D_{NC}=Empirical function of IWC,R_{NEW} $\lambda=(E/D_{NC})R_{NEW}$

D_{min}=0.08mm (Field and Heymsfield, 2003)

Old precip grows by riming, removes ice-soluble species

$\lambda=(E/D_{OCA})R_{OCA}$

If R(L)-R_{AM}-R_{OCA}-$GROWTH_{ICE}$>0, new precip forms as above

A single D falls into cloud below

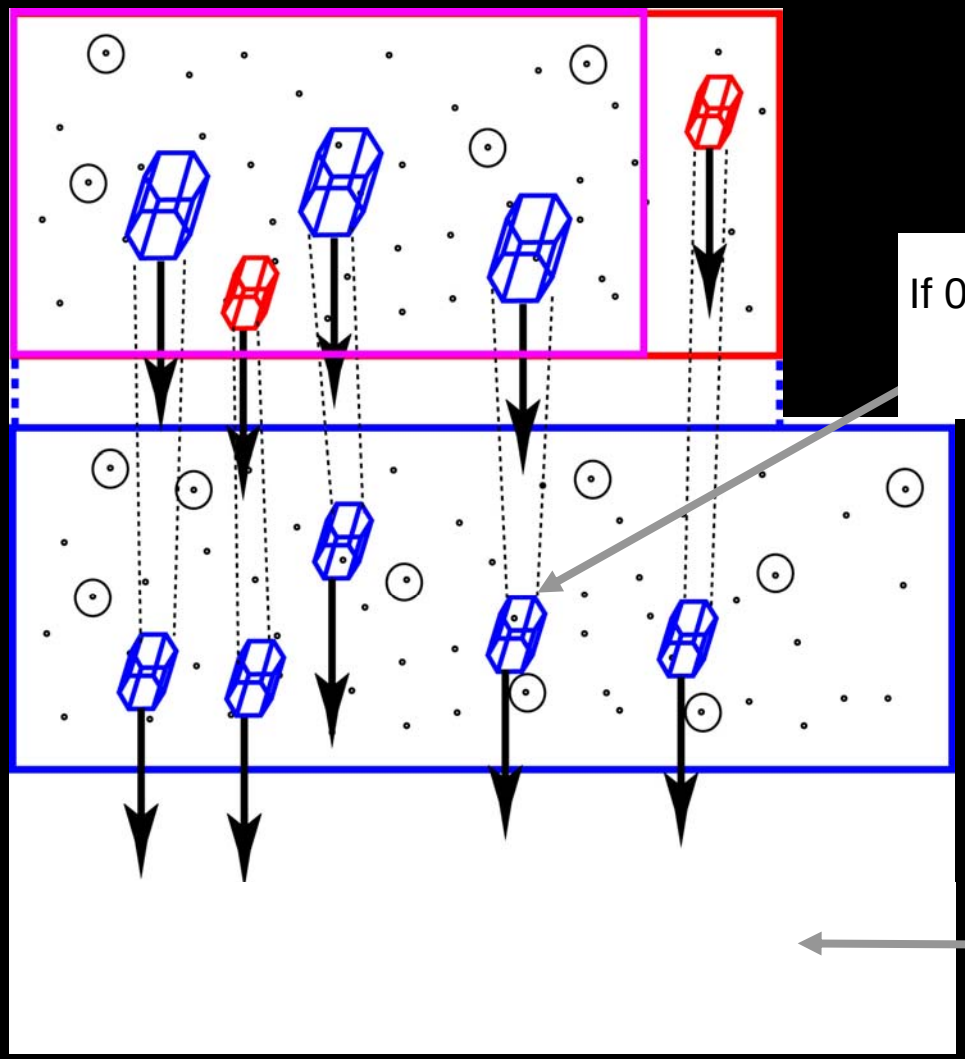
$D=(F_1*D_{OC} + F_2*D_{NC})/CF(L-1)$

$D_{OC}=f(IWC_{OC},R_{OCA}+R_{RIME}+R_{NEWOC})$

(Field and Heymsfield, 2003)

D=const in Ambient

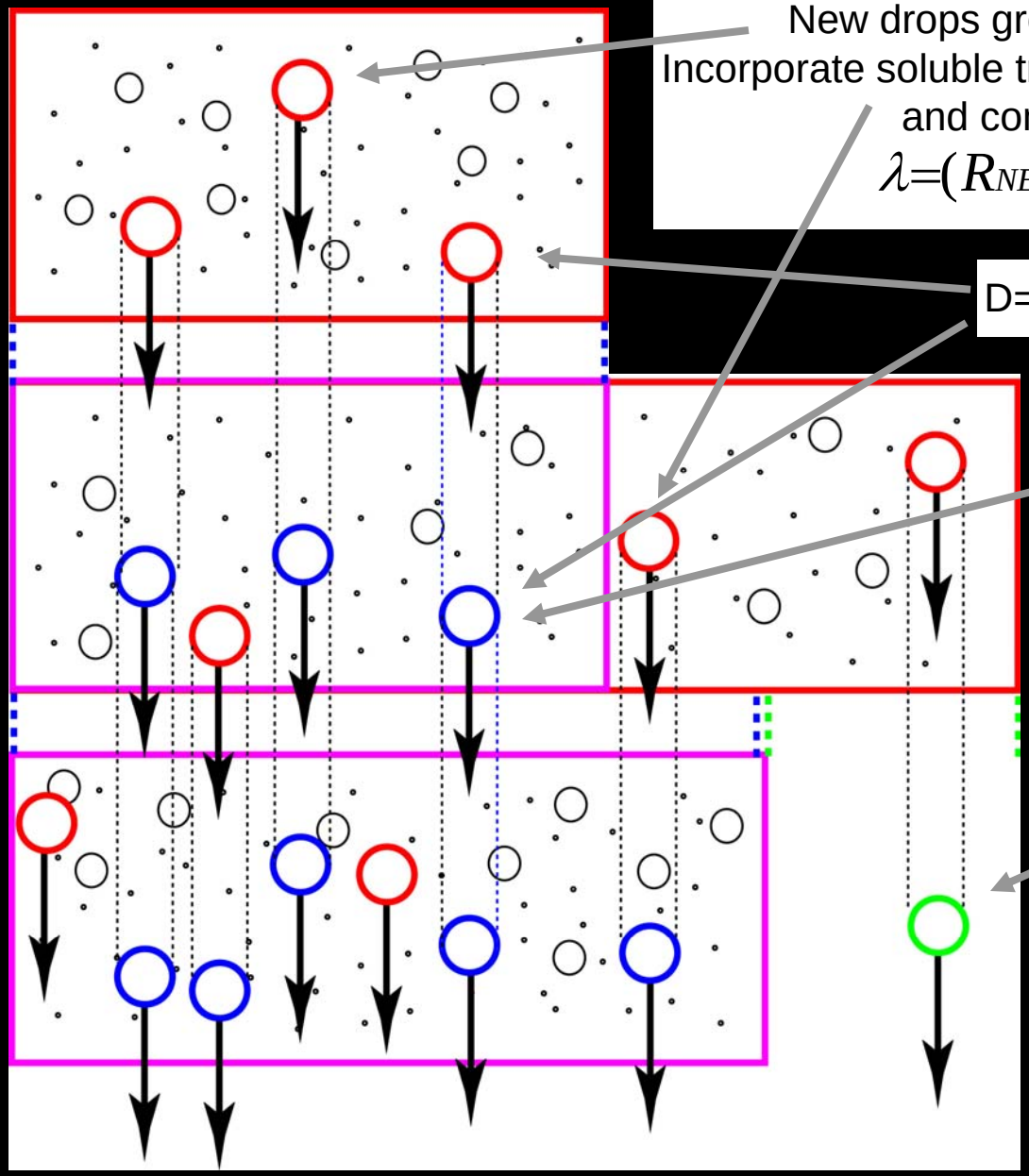
No washout since gases are in vapor phase Evaporation at a constant rate



If 0<R(L-1)<R(L), decrease the ice particle size

No tracer evaporation or removal

If R=0, evaporate all tracer from ice



New drops grow by collision and coalescence
Incorporate soluble tracers within CF based on Henry's Law and conversion of CLW to precip

$$\lambda = (R_{NEW} H^*/F)(0.29 P/M_{AIR})$$

D=10mm for all rain

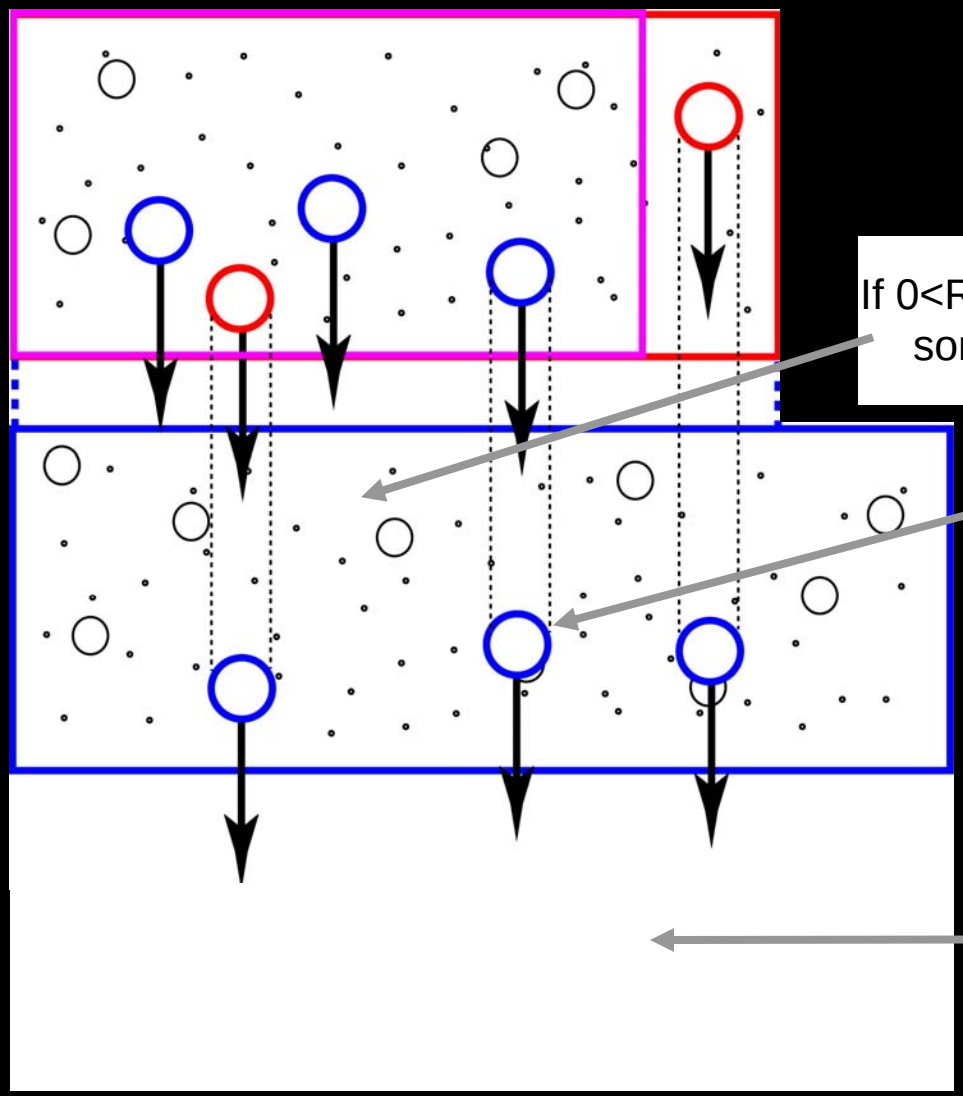
Scavenging by old precip Henry's Law limited for moderately soluble species, mass transfer-limited for highly soluble species

$$\lambda = (R_{OCA} H^*/F)(0.29 P/M_{AIR})$$

or

$$\lambda = (E/D_R) R_{OCA}$$

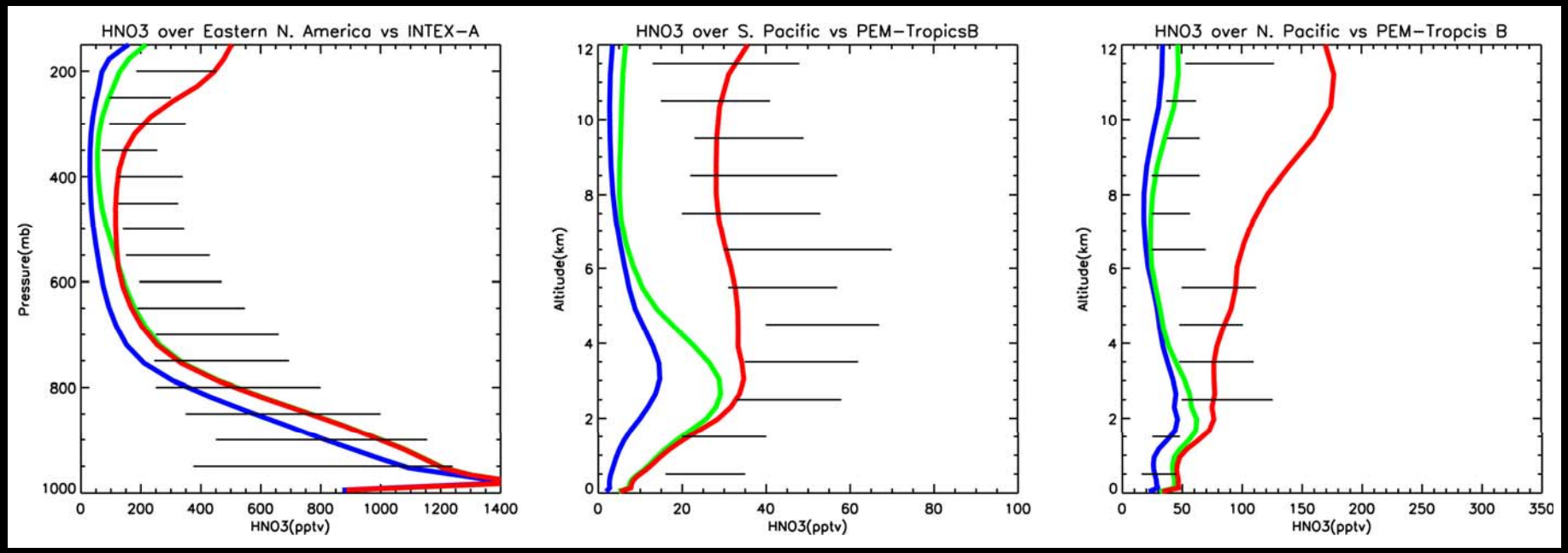
Scavenging in Ambient
(Henry's Law or Kinetically limited)



If 0<R(L-1)<R(L), assume complete evaporation of some raindrops and release of soluble gases

Remaining raindrops scavenge (Henry's Law or Kinetically limited)

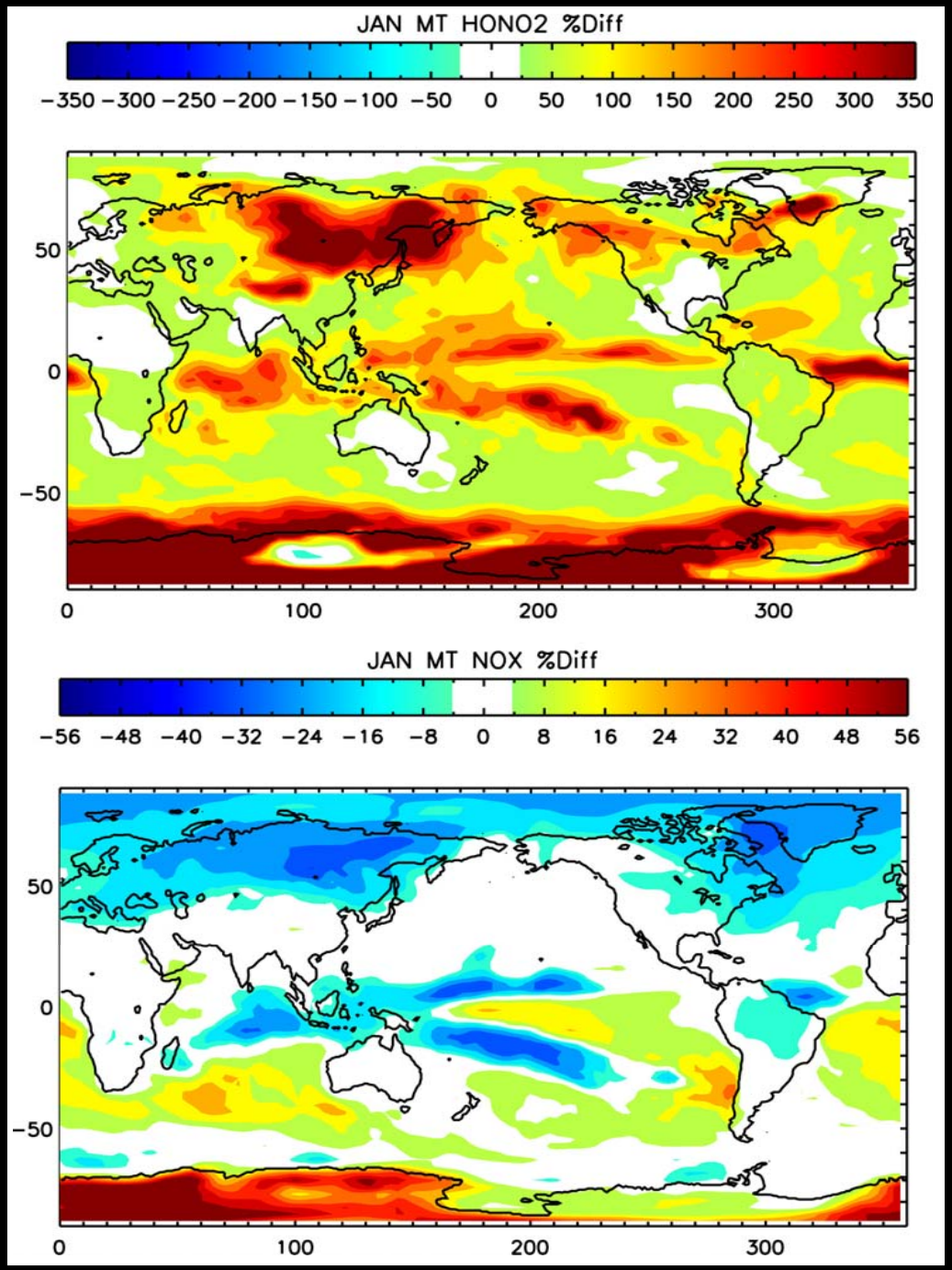
If R=0, evaporate all tracer



$SCAV_{ice} = SCAV_{Liquid}$ No Cloud Overlap

$SCAV_{ice} = SCAV_{Liquid}$ Cloud Overlap

$SCAV_{ice}$ = Impaction and Riming, Cloud Overlap



Much of the geographical distribution comes from cloud overlap. Ice has largest impact at high latitudes and in the upper troposphere

Indirect impact on other species not straightforward – e.g. HONO2 vs H2O2

Summary

The cloud overlap scheme proposed here is one way to carefully partition precipitation scavenging into in-cloud and below-cloud removal.

Cloud overlap increases the abundance of soluble species in the boundary layer and mid-troposphere – can we explain the geographical patterns?

Ice grows into precipitation in a fundamentally different way than rain – it may not be reasonable to use classic “rainout” formulations for ice

Using an impact scavenging formulation for ice formation results in much less HONO2 removal in the upper troposphere and high latitudes

Mixed-phase clouds are an open question – high latitude measurements will be key