## A Decade of Polar Stratospheric Cloud Observations from CALIOP

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### What are Polar Stratospheric Clouds (PSCs) and why are they important?

- PSCs form in the Antarctic and Arctic stratosphere at temperatures below ~195 K
- Three known particle compositions: liquid super-cooled ternary solution (STS) H<sub>2</sub>SO<sub>4</sub>-HNO<sub>3</sub>-H<sub>2</sub>O droplets, nitric acid trihydrate (NAT) crystals, and H<sub>2</sub>Q



- acid trihydrate (NAT) crystals, and H<sub>2</sub>O PSCs play key roles in chemical depletion of ozone at high latitudes Ice
  - Heterogeneous (mixed-phase) reactions on PSC particle surfaces convert inactive chlorine reservoirs (HCl and ClONO<sub>2</sub>) to active (ozone-depleting) chlorine radicals
  - Sedimentation of large NAT particles irreversibly removes odd nitrogen (denitrification), which delays chlorine deactivation and prolongs ozone depletion
- Significant gaps in knowledge still exist
  - Large solid particle formation and their denitrification potential (NAT rocks)
  - Extent of chlorine activation on cold background stratospheric aerosol
  - Limit our ability to accurately represent PSCs in global models and call into



### CALIOP has greatly expanded our PSC database

Typical Daily Antarctic Winter Coverage 2008/07/17 (blue=night, red=day)



- Historical PSC database rather sparse
- Extensive measurement coverage over polar regions into polar night
- High spatial resolution (5-km horizontal x 180-m vertical resolution PSC product)
- Combination of total backscatter and polarization sensitive measurements provide information on PSC

532 nnSATWithil Attenmated Balas A Des kto 2017-2008-07-17





### **PSC Detection and Composition Classification** Pitts et al., ACP, 2007; 2009; 2011; 2013

- PSCs are detected as statistical outliers from background aerosol using nighttime 532-nm scattering ratio (total/molecular backscatter, R<sub>532</sub>) and perpendicular backscatter, β<sub>⊥</sub>
- Composition classification based on comparison of CALIOP particle depolarization ratio  $\delta_P$  and inverse scattering ratio  $1/R_{532}$  observations with theoretical optical calculations
  - PSCs separated into five composition classes
  - $\succ$  β⊥ outliers: NAT mixtures/ice; R<sub>532</sub> outliers: STS
  - Boundary between Mix2-enh and ice is also adjusted to account for effects of denitrification and dehydration
- CALIPSO Level 2 PSC data product available from Langley Atmospheric Sciences Data Center:





- \$STS = supercooled ternary (H<sub>2</sub>SO<sub>4</sub>-H<sub>2</sub>O-HNO<sub>3</sub>)
  solution
- \*<u>Mix1+Mix2</u>, Mix 2-enh(anced) = external mixtures of liquid (binary  $H_2SO_4$  aerosol or STS) droplets and NAT particles (in increasing number density)
- $let let let H_2O$  ice (synoptic, mountain-waveinduced)



### CALIOP PSC Composition Classification 17 July 2008

532-nm Backscatter Ratio



#### 532-nm Perpendicular Backscatter



PSC Composition





# CALIOP Antarctic PSC Climatology 2006–2015

#### Antarctic PSC Areas: 2006–2015





#### Antarctic PSC Area Fraction by Composition Vortex Average: 2006–2015



- STS is predominant composition early and again late in the season
- Mix1+Mix2 are predominant at the lowest altitudes throughout the season
- Ice PSCs are never the primary composition and typically are episodic
- Relative maximum in Mix2-enh PSCs above ~ 20 km during July-August period



#### Monthly Average Spatial Distributions (2006–2015) Antarctic PSC state 20 km altitude



**PSC** Frequency



# CALIOP Arctic PSC Observations 2006–2016





#### Monthly Average Spatial Distributions (2006–2015) Arctic PSC anti-200 km altitude





#### CALIPSO/microphysical modeling studies have revised our understanding of PSC HJSO4/HJO nucleation





# Accepted nucleation pathways

- Growth of liquid particles due to uptake of HNO<sub>3</sub> (Dye et al., 1992; Carslaw et al.,1994)
- 2) Homogeneous nucleation of ice particles (Koop et al., 2000)
- NAT nucleation on pre-existing ice particles (Carslaw et al., 1998)
- Widespread NAT PSCs observed by CALIPSO in Arctic during December 2009
- No ice PSCs observed and meteorological conditions unfavorable for ice formation
  - NAT formed before ice PSCs were present- what was the mechanism?

# **PSC Formation: New Heterogeneous Pathway**

M2

M1

1e-7



Heterogeneous ice and NAT nucleation on foreign nuclei imbedded in LSA (immersion freezing)

Evidence for the existence of foreign nuclei (e.g. Weigel et al., 2014)

Parameterization based on active site theory (Marcolli et al., 2007)

> Heterogeneous nucleation on pre-existing solid particles (not ice) required to explain CALIOP NAT observations in December 2009

Hoyle et al., *Atmos. Chem. Phys., 13*, 9577–9595, 2013.



# **PSC Formation: New Heterogeneous Pathway**



Heterogeneous ice and NAT nucleation on foreign nuclei imbedded in LSA (immersion freezing)

Evidence for the existence of foreign nuclei (e.g. Weigel et al., 2014)

Parameterization based on active site theory (Marcolli et al., 2007)

M2

1e-5

Heterogeneous nucleation on pre-existing solid particles <u>plus</u> small-scale temperature fluctuations required to explain CALIOP synoptic ice observations in January 2009

Engel et al., *Atmos. Chem. Phys.,* 13, 10769–10785, 2013.



H<sub>2</sub>SO<sub>4</sub> / H<sub>2</sub>O

liquid sulfate aerosol

T [K]

# Orographic and Tropospheric Forcing of PSCs



 Association of PSC formation with underlying upper tropospheric clouds and blocking anti-cyclones (Wang et al., JGR 2008; Adikhari et al., JGR 2010; <u>Kohma al</u>

 Influence of orographic gravity waves (OGW) on PSC production (Noel et al., JGR 2009; <u>Alexander et</u> <u>al., JGR 2011</u>, JGR 2013; Noel and Pitts, JGR 2012)



### A-Train Synergy: Observations of Vortex-wide Chlorine Activation by Mesoscale PSC Event



Mesoscale PSC events in early winter can rapidly activate chlorine in just a few hours and effectively activate the whole polar vortex in a few days Wegner et al., Atmos. Chem. Phys., 16, 4569-4577,



# Polar Stratospheric Cloud



*initiative* http://www.sparc-climate.org/activities/polar-stratosphericclouds/

- PSCi began as a new SPARC activity in November 2015
  - Leads: Michael Pitts, Ines Tritscher, Lamont Poole, and Thomas Peter
- Main objectives:
  - Assess recent research developments related to PSCs
  - Compare remote and in situ datasets to identify their strengths and limitations
  - Identify the key PSC characteristics required by global models that can be estimated from measurements
  - Synthesize new datasets into a state of the art PSC climatology
  - Identify remaining open science questions
- Will ultimately lead to improved representation of PSC processes in global climate models
- Deliverables
  - Reference PSC data records
  - Comprehensive review paper on state of PSC science



# Summary and Outlook

- CALIOP is providing a wealth of information on PSC occurrence and composition on unprecedented spatial scales
- CALIOP 10-year data record has captured primary aspects of the seasonal and multi-year variability of PSCs in Antarctic and Arctic
  - Small interannual variability in Antarctic: Multi-year averages fairly representative
  - > Large interannual variability in Arctic: Each Arctic winter is unique
- CALIPSO/RECONCILE partnership has led to an improved understanding of PSC processes (POLSTRACC studies in early stage)
  - CALIPSO observations provided vortex-wide and season-long context to the focused RECONCILE campaign
  - CALIPSO/microphysical modeling studies have revised our understanding of NAT and ice nucleation
- Next major steps:
  - Development of detailed PSC reference climatology and review paper based on CALIOP, MIPAS, MLS and other datasets (SPARC PSC initiative)



# BACKUP SLIDES



# CALIPSO played significant role in European Arctic field campaigns

- Invited to participate as Associated Partners in RECONCILE (EU) and POLSTRACC (Germany) projects
- CALIOP quick-look images used to identify PSC regions for flight planning purposes
- Provided overall context to PSC season (Arctic-wide view of PSCs)
- Coordinated under-flights of CALIPSO with Geophysica and HALO research aircraft
- Quick-look comparison of CALIOP PSC data products with aircraft and balloon-borne data during field mission
- Partnerships have led to an improved understanding
   If PSC processes





#### Hemispheric Differences in Composition



- >15 times more PSC observations in the Antarctic
- Year-to-year variability in PSC composition much higher in the Arctic
- Fraction of ice PSCs is a factor of nine smaller in the Arctic
- Fraction of Mix 2-enh PSCs is more than a factor of two smaller in the Arctic