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Key Points:

- Large (5 microns) ash particles detected in the tropical lowermost stratosphere
- Ash particles were from the Mount Kelud eruption and persisted in the stratosphere for 4 weeks
- The low ash particle concentration indicates a limited impact on ice clouds or radiation

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Ash Particles Detected in the Tropical Lower Stratosphere

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Abstract We report here on measurements of relatively large (up to ≃5-μm maximum dimension) particles detected in the tropical western Pacific lower stratosphere. The particles were observed at low relative humidities and contain no detectable ice mass. We conclude that these particles are silicate ash injected by the Mount Kelud eruption in Indonesia a few weeks prior to the aircraft observations. The Kelud ash particles were detected by cloud probes on the National Aeronautics and Space Administration Global Hawk between the lapse rate tropopause (≃16.5 km) and the aircraft ceiling (18.5 km). The ash particles detected in the lowermost stratosphere by the aircraft extended well into the northern subtropics. The concentration of ash particles dropped off rapidly below the lapse rate tropopause, presumably because of rapid removal by ice nucleation scavenging. Although ash particles are very effective ice nuclei, the low particle concentrations detected in the upper troposphere precludes a significant impact on cirrus clouds.

Plain Language Summary We report here on measurements of relatively large (up to 5-μm maximum dimension) particles detected in the tropical western Pacific lower stratosphere during the National Aeronautics and Space Administration Airborne Tropical TRopopause EXperiment Global Hawk flights in early March 2014. The particles were observed at low relative humidities and contain no detectable ice mass. We conclude that these particles are silicate ash injected by the Mount Kelud volcanic eruption in Indonesia a few weeks prior to the Airborne Tropical TRopopause EXperiment flights. The Kelud ash particles were detected by cloud probes on the Global Hawk between the lapse rate tropopause (16.5 km) and the aircraft ceiling (18.5 km). The ash particles sampled in the lowermost stratosphere by the aircraft extended further north than the Kelud plume measured by spaceborne lidar at higher altitudes. The concentration of ash particles dropped off rapidly below the lapse rate tropopause, presumably because of rapid removal by ice nucleation on the ash particles followed by ice crystal sedimentation. Although ash particles are very effective ice nuclei, the low particle concentrations detected in the upper troposphere preclude a significant impact on cirrus microphysical properties.

1. Introduction

Explosive volcanoes that inject gas phase and particulate matter into the stratosphere have large impacts on interannual and decadal climate variability (Fyfe et al., 2013; Solomon et al., 2011). Most modeling studies focus on the volatile sulfate aerosols resulting from the volcanic sulfur injections, but silicate ash particles can also be injected into the stratosphere, and they can be an important component of the volcanic plume optical properties, at least during the first few weeks after the eruption (Oberbeck et al., 1983; Turco et al., 1983; Vernier et al., 2016). High-altitude measurements after the eruptions of Mount St. Helens in 1980 and El Chichon in 1982 showed that a broad size distribution of ash particles can be injected into the stratosphere, with particles as large as 30–40 μm (Farlow et al., 1981; Gooding et al., 1983). The largest particles fall out of the stratosphere rapidly, but ash particles with maximum dimensions of a few microns persisted up to 6 months after the El Chichon eruption, possibly suggesting particle sedimentation speeds were slowed by irregular shapes and low-density clusters of particles (Gooding et al., 1983). Numerical simulations indicate

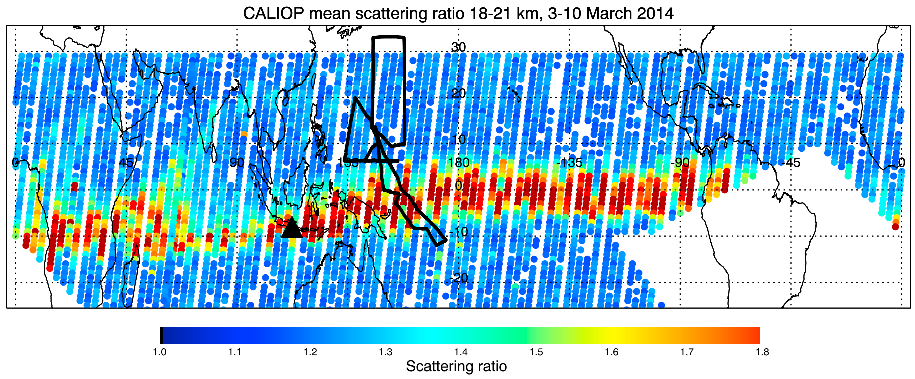

Figure 1. Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) scattering ratio averaged over 3–10 March and 18–21 km. The location of the Mount Kelud eruption in Indonesia is indicated by the black triangle. The Airborne Tropical TRopopause EXperiment flight paths from Guam are shown in black for reference. In this altitude range, the Kelud stratospheric plume has spread throughout the tropics, but is mostly south of 5°N.

that particles larger than about 5-μm diameter will be removed from the stratosphere by sedimentation within a few weeks (Turco et al., 1983).

As documented in detail by Vernier et al. (2016), the eruption of Mount Kelud (Indonesia) on 13 February 2014 resulted in a persistent aerosol plume in the tropical lower stratosphere. Backscatter and depolarization measurements from the Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) on board Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations satellite were used to investigate the evolution of the Kelud ash and sulfate particles in the lower stratosphere. The stratospheric aerosol plume spread throughout the global tropics within a few weeks (Figure 1), and lower stratosphere aerosol extinction remained well above background levels for at least a few months. Balloon-borne backscatter sondes launched from northern Australia in May 2014 indicated that fine ash (radii less than 0.3 μm) persisted in the lower stratosphere 3 months after the eruption (Vernier et al., 2016). The sulfate aerosols and ash particles became vertically separated with time owing to faster sedimentation speeds of the larger ash particles. The sulfate aerosol layer remained between about 18 and 22 km, whereas the ash particles were mostly below 20 km. Note that the CALIOP analysis presented by Vernier et al. (2016) was restricted to altitudes above 18 km because cirrus occurring at lower altitudes can dominate the backscatter measurements.

During mid-February through early March 2014, the National Aeronautics and Space Administration Airborne Tropical TRopopause EXperiment (ATTREX) conducted flights from Guam with the long-range Global Hawk aircraft. The science foci of ATTREX were tropical tropopause layer (TTL; ≃14–18 km) transport, chemistry, cloud formation, and dehydration. The ATTREX Global Hawk payload provided measurements of water vapor, meteorological conditions, cloud properties, tracer and chemical radical concentrations, and radiative fluxes (Jensen et al., 2017). Although the ATTREX science team was aware of the Kelud eruption, flights were not specifically designed to target the aerosol plume because of a lack of aerosol instrumentation on the aircraft. However, as we will show in this paper, the Kelud ash particles in the lowermost stratosphere were detected by the cloud probes on the Global Hawk during the early March flights in the western Pacific Northern Hemisphere tropics. The in situ aircraft measurements provide information about particle size and concentration that is complementary to the CALIOP measurements.

2. ATTREX Measurements

The key instrument used to detect the Kelud ash particles was the Fast Cloud Droplet Probe (FCDP; Lawson et al., 2017; McFarquhar et al., 2007; Woods et al., 2018), which measures the forward scattered light from individual particles. Mie scattering calculations are used to retrieve particle sizes from 1- to 50-μm diameter. Without sufficient sulfate uptake, ash particles would be nonspherical, resulting in an FCDP sizing uncertainty of up to about a factor of 2 (Borrmann et al., 2000). The FCDP is an open-path instrument designed for sampling cloud droplets and ice crystals. The relatively low sample volume of FCDP (0.08 L/s; Thornberry et al., 2017) results in a minimum detectable particle concentration in 1-s samples of about 15 L^{-1}. Since the lower stratospheric ash particle concentration was relatively low, we use 15- and 60-s averages of FCDP data in our analysis to obtain sufficient statistics for determination of particle size distributions and concentrations.

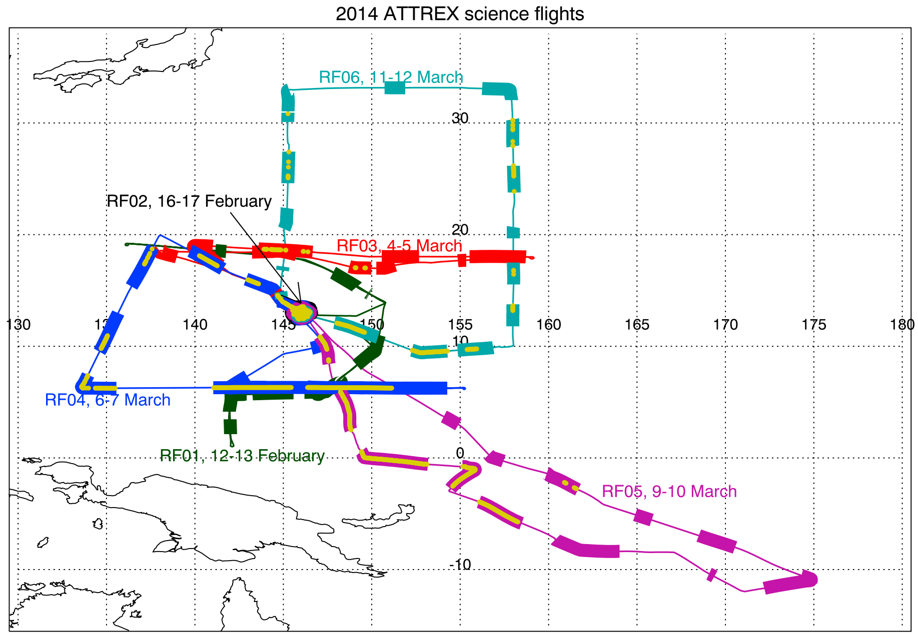

Figure 2. Airborne Tropical TRopopause EXperiment (ATTREX) February–March 2014 flight tracks from Guam. The thick sections of the flight tracks indicate where the aircraft was above the local lapse rate tropopause height (16.5 km on average; see text for discussion of tropopause height determination). Yellow dots along the flight paths indicate locations where 15-s samples indicated particles present (detected by Fast Cloud Droplet Probe), relative humidity with respect to ice < 50%, and no ice detected by NOAA-H_2O. These particles were primarily detected when the aircraft was in the lowermost stratosphere during the last three flights (6–12 March).

The Global Hawk payload included two FCDP probes, one a stand-alone FCDP, and the other as part of the Hawkeye probe. The agreement between the ash particle properties indicated by the two FCDP instruments provides additional confidence that the particle detections are not instrument artifacts.

Important complementary measurements used to distinguish ash particles from ice crystals are temperature, pressure, water vapor concentration, and total water concentration. Temperature and pressure were measured by the Meteorological Measurement System, with a temperature uncertainty of 0.3 K. Water vapor was measured with both external-path and internal-path diode laser hygrometers. The two measurements showed excellent agreement even under the very dry conditions in the tropical lower stratosphere (Jensen et al., 2017). Here we use the internal-path NOAA-H_2O water vapor measurement, which has an uncertainty of 5% $\pm$0.23 ppmv (Thornberry et al., 2015). The resulting uncertainty in calculated relative humidity with respect to ice (RHI) is no more than 15%.

The NOAA-H_2O instrument also includes a total water measurement using a forward-facing inlet (Thornberry et al., 2015). Ice crystals are inertially enhanced by a factor ranging from 33 to 48, depending on the particle size. The cloud ice water content (IWC) is obtained by differencing the total water and vapor channels, and the inertial enhancement provides a high sensitivity of about 2–3 $\mu g/m^3$ (corresponding to about 0.02–0.03 ppmv at 100 hPa and 200 K; Thornberry et al., 2017). The ability to detect small amounts of ice present provides a sensitive discrimination between ash particle layers and ice clouds.

The six ATTREX Global Hawk flights from Guam covered the western Pacific region from about 10°S to about 30°N (Figure 2). The sampling strategy used most of the time was to profile between about 14 km and the Global Hawk ceiling, which varied between about 16 and 18.5 km, depending on the fuel load. This approach provided numerous vertical profiles through the TTL.

3. Observations of Ash Particles From Kelud Eruption

The central science focus of ATTREX was ice cloud processes controlling water vapor concentration in the TTL and in air entering the stratosphere. Examination of the RHI frequency distribution inside clouds is a useful method for evaluating numerical models of cirrus formation and interactions with water vapor. The RHI

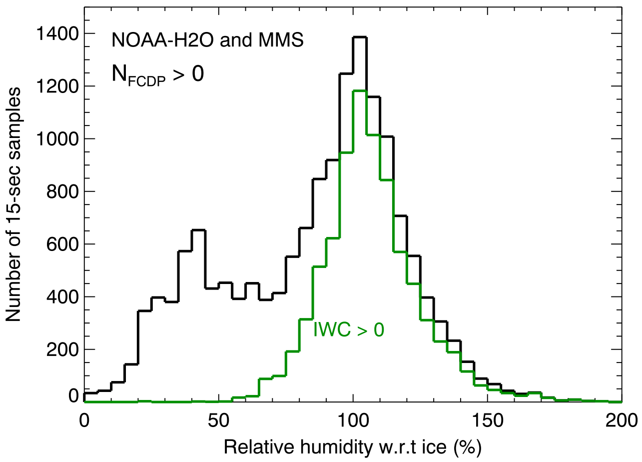

Figure 3. Relative humidity frequency distributions during 15-s samples when Fast Cloud Droplet Probe (FCDP) detected particles. Black curve: All samples with FCDP particles; green curve: only including samples with condensed ice detected by the NOAA-H_2O instrument. The mode at relative humidity with respect to ice (RHI) < 50% corresponds to the stratospheric ash particles.

frequency distribution including only 15-s samples where the FCDP probe detected particles (Figure 3) has the expected peak near 100% and tail extending to large supersaturations. However, a second peak is apparent at RHI less than 50%. This peak is almost exclusively produced by measurements made in the lower stratosphere. The possibility that these particles are sublimating ice crystals is implausible since small (diameters less than 10 μm) ice crystals should sublimate within a few minutes at RHI < 50%. Further, if we include only samples with ice detected by the NOAA-H_2O IWC measurement, then the peak at low RHI disappears (Figure 3, green curve), indicating that the particles detected at low relative humidity are not ice crystals. All of the observational data used in this study are publicly available on the Earth Science Project Office archive (espoarchive.nasa.gov).

Size distributions of the ash particles sampled in the lower stratosphere are shown in Figure 4. Size distributions from the two FCDP probes on the aircraft agree well. The particle maximum dimensions indicated by FCDP ranged from the lower detection limit (0.5 μm) up to about 10 μm; most of the particles had maximum dimensions less than about 5 μm. These particles are much too large to be sulfate aerosols. The fall speed of a 3-μm diameter spherical particle with a density of 2.5 g/cm^3 under lower stratospheric conditions is about 0.1 cm/s, which implies a sedimentation distance of about 3 km over the 3–4 weeks since the Kelud eruption. If the ash particles are nonspherical, they would fall at somewhat slower speeds. Likewise, if the ash particles are coated with sulfate, the density would be lower, resulting in slower sedimentation. During the first 10 days after the eruption, ash particles were detected by CALIOP in a primary layer between 18 and 20 km, with a secondary layer at about 22–23 km (Vernier et al., 2016). Therefore, it is plausible that the particles detected between ≃16.5 and 18.5 km during ATTREX were ash from the Kelud eruption.

The FCDP measurements also provide information about the lower stratospheric ash particle concentration. We use 60-s averages here to allow detection of low particle concentrations, but this involves averaging over about 10 km of horizontal distance, and we thereby lose information about variability in the particle concentrations at smaller scales. The FCDP measurements indicate lower stratospheric particle concentrations less than 10 L^{-1}. Even with 10-km samples, the ash particle concentration is highly variable along the flight tracks (see Figure 2). The ash extinctions at visible wavelengths indicated by the FCDP measurements are about 0.0001–0.0003 km^{-1}, which is comparable to the background aerosol extinction in the tropical lower stratosphere (Thomason & Vernier, 2013).

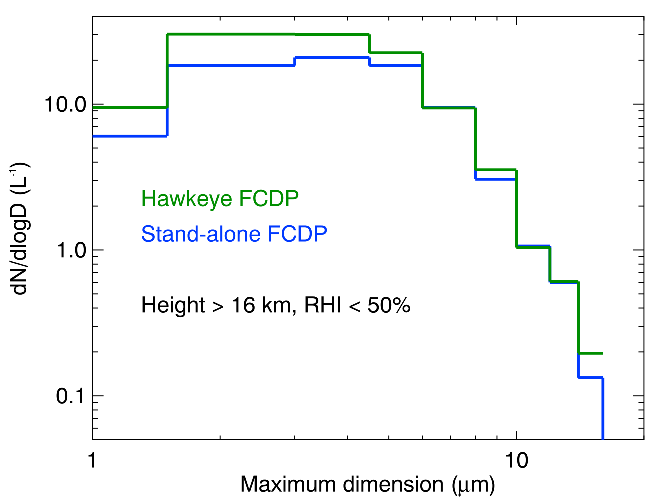

Figure 4. Stratospheric ash particle size distribution. Only samples above 16.5 km and with relative humidity with respect to ice (RHI) < 50% are included. The two curves show the size distributions indicated by the two Fast Cloud Droplet Probe (FCDP) probes on the Global Hawk flights from Guam.

The relative humidities corresponding to samples with ash particles and ice crystals are plotted versus height relative to the lapse rate tropopause in Figure 5. For Global Hawk vertical profiles that extended into the lower stratosphere, the lapse rate tropopause is identified using a modified World Meteorological Organization definition (World Meteorological Organization, 1957), where we reduced the depth of the layer in which the average lapse rate must be 2 K/km from 2 to 0.5 km (Pan et al., 2018). For aircraft profiles that did not extend into the stratosphere, we use nearby (within 12 hr and 500 km) radiosonde or COSMIC profiles. The data from the few profiles with none of these options available are not included. Ash particles (red points) correspond to 15-s samples with FCDP detection of particles but no ice mass indicated by the NOAA-H_2O IWC measurement. The green points show samples with ice detected (cirrus clouds). Occurrence frequency height distributions (solid curves in Figure 5) show that ice crystals were almost exclusively confined to heights below the lapse rate tropopause, whereas the ash was mostly detected above the tropopause. The concentrations of ash particles were correspondingly much lower in the upper troposphere than in the stratosphere. Also, Figure 5 shows

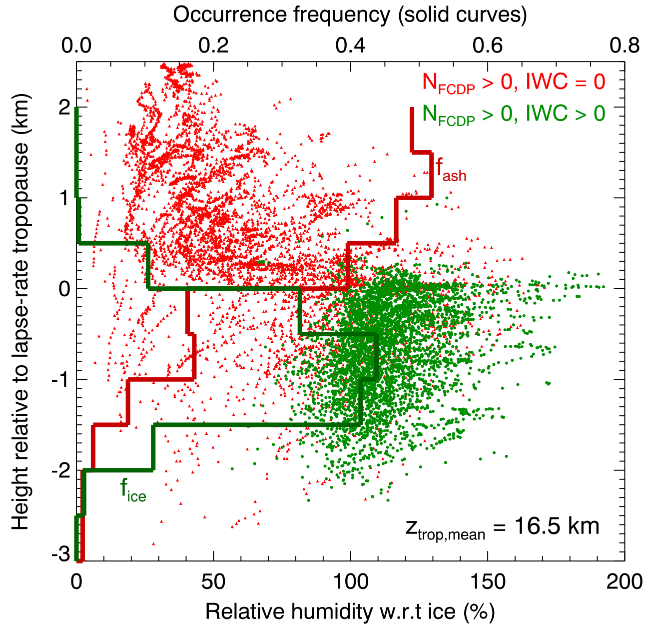

Figure 5. Height distributions of relative humidity with respect to ice relative to the lapse rate tropopause of 15-s samples with ice crystals (green circles, defined as particles detected by FCDP and IWC detected by NOAA-H_2O) and ash particles (red triangles, defined as particles detected by FCDP and no IWC). The solid lines show the fractions of 15-s samples with ice or ash particles detected. See text for discussion of lapse rate tropopause height determination. FCDP = Fast Cloud Droplet Probe; IWC = ice water content.

expected clear distinction between low relative humidities where the ash particles were detected above the tropopause and high relative humidities in the ice clouds below the tropopause.

Although the ash particle concentration drops off sharply below the tropopause, some ash particles are detected in dry regions in the upper troposphere. Removal of ash particles in the lower stratosphere is controlled by horizontal dispersion and sedimentation, with corresponding long lifetimes. In the upper troposphere, ash particles will be more rapidly removed by overturning circulations and ice nucleation scavenging (see discussion below). The difference in ash concentration between the lower stratosphere and upper troposphere may just be a result of the removal lifetime difference.

The CALIOP measurements at altitudes above 18 km indicated that the Kelud stratospheric aerosol enhancement was confined to latitudes south of 5°N in the ATTREX sampling time period and longitude range (Figure 1). In contrast, the ATTREX observations indicated the lower stratospheric ash layer extended well into the northern subtropics (Figure 2). However, the CALIOP analysis was confined to altitudes above 18 km. The more rapid poleward transport in the lowermost stratosphere is consistent with the known height variation in stratospheric latitudinal mixing and advection.

To understand the differences between the broad northward extent of the Kelud ash cloud at 16.5–18.5 km implied by Figure 2, and the latitudinally compact distribution above 18 km (Figure 1), we launched 30-day forward trajectories from a 3° by 3° region around Kelud at times bracketing the eruption at 16 Z, 13 February 2014. We used half-degree 6-hourly ERA-Interim analyses, carefully interpolated to very high vertical resolution in the TTL using the technique documented by Kim and Alexander (2013). Diabatic heating was radiative (including clouds), as determined by Yang et al. (2010). The results are in Figure 6, showing the positions of all parcels on 3–13 February inclusive, color coded by age. Above 18.5 km, the parcels are tightly confined, with some spread between 18 and 18.5 km.

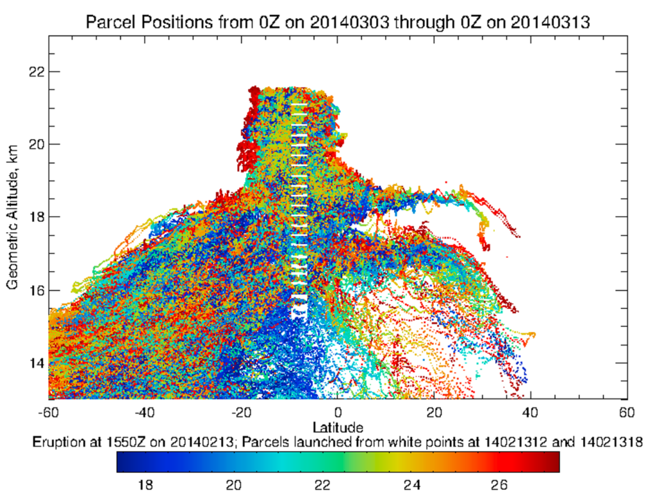

Figure 6. Latitudes and heights of trajectories launched from the Kelud eruption location and time. Colors indicate the time since the eruption. White bars indicate the trajectory launch locations. The rapid poleward transport of parcels below about 18.5 km is evident. The Cloud-Aerosol Lidar with Orthogonal Polarization observations that were limited to heights above 18 km indicated ash particles only south of about 7°N (Figure 1), whereas ash particles were detected by the aircraft at altitudes below 18.5 km as far north as about 30°N (Figure 2).

The Northern Hemisphere TTL monsoon anticyclone near 17 km spreads some parcels northward as far as the subtropical jet. The Austral summer anticyclones spread parcels southward; the relatively weak summer subtropical jet is a less effective barrier to latitudinal transport than during wintertime. Note that there is some northward spread between 18 and 19 km. However, only about 3% of the parcel positions are north of 10°N in that altitude range (vs. 11% between 16.5 and 18 km).

We also examined data from specific CALIOP orbits in the ATTREX sampling region and time period. Aerosol enhancements in the lowermost stratosphere (between about 16.5 and 18 km) were not apparent. However, as noted above, the extinction associated with the ATTREX FCDP ash measurements is comparable to or lower than the background aerosol extinction; therefore, detection of these particles by CALIOP would not necessarily be expected, and the fact that the Kelud ash plume in the lowermost stratosphere is not detected by CALIOP does not indicate an inconsistency in the measurements. Essentially, the CALIOP and aircraft measurements provide complementary information about the Kelud plume: The aircraft measurements provide more sensitive detection of ash particles in the lowermost stratosphere than is possible with spaceborne lidar, whereas the CALIOP measurements provide global coverage over the entire evolution of the plume.

4. Summary and Discussion

We have presented measurements from the 2014 ATTREX Global Hawk flights in the tropical western Pacific that provided unexpected evidence of relatively large volcanic ash particles in the lowermost stratosphere. The key results are summarized as follows:

1. A widespread layer of relatively large (maximum dimensions up to about 5 μm) particles was detected in the tropical western Pacific lowermost stratosphere (between about 16.5 and 18.5 km) at relative humidities less than 50% on the ATTREX Global Hawk flights in early March 2014.
2. These particles contained no detectable ice mass and were presumably ash particles from the Mount Kelud eruption a few weeks earlier (13 February 2014). This conclusion is consistent with the CALIOP observations of the Kelud injection of ash well into the stratosphere as well as sedimentation speeds of 3-μm particles in the lower stratosphere. The aircraft measurements below 18.5 km indicate ash particles at higher northern latitudes than the CALIOP measurements above 18 km. This difference in latitudinal extent of the ash cloud at different altitudes is consistent with the height dependence in transport to the extratropics (Figure 6).
3. The measured concentrations of ash particles in the lower stratosphere are about 1–10 L^{-1}. Extinctions calculated from the particle size and concentration measurements are comparable to the background aerosol extinction.
4. The concentration of particles that contain no detectable condensed water (i.e., not ice crystals) decreased sharply below the lapse rate tropopause (16.5 km on average). This change in concentration is presumably a result of rapid removal of the ash particles in the upper troposphere by nucleation scavenging and convective overturning.

Ash particles can be very effective ice nuclei (Schill et al., 2015). As a result, they can promote formation of cirrus clouds at lower relative humidities than is possible with only homogeneous nucleation or nucleation on less effective ice nuclei. However, the concentrations of ash particles in the lower stratosphere (no more than 1–10 L^{-1}) is relatively low, and the ash particle concentration drops rapidly below the tropopause where cirrus formation is possible. Cirrus with concentrations less than 10 L^{-1} have very low optical depths, and growth of ice crystals in such low ice concentration cirrus will not appreciably deplete the vapor in excess of saturation (Jensen et al., 2013). In a steadily cooling air parcel, the relative humidity will continue to rise after nucleation on the few ash particles until additional ice crystals are generated by heterogeneous nucleation on more abundant, less effective ice nuclei (such as crystalline ammonium sulfate or glassy organic particles; Jensen et al., 2018) or by homogeneous freezing of aqueous aerosols. The resulting cloud microphysical and optical properties will be dominated by the less effective ice nuclei or homogeneous freezing. However, IN concentrations of 10 L^{-1} have very little impact on the balance between heterogeneous and homogeneous ice nucleation. Significantly larger IN concentrations are required to alter cirrus microphysical properties. Overall, nucleation of ice crystals on the few Kelud ash particles present during the ATTREX flights probably did not significantly alter TTL cirrus microphysical properties or the water vapor budget. However, in the fresh ash plume just after the eruption, ash particle concentrations may have been high enough for significant impacts on cirrus.

Acknowledgments
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