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# Overshooting of clean tropospheric air in the tropical lower stratosphere as seen by the CALIPSO lidar

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Received: 12 August 2010 – Accepted: 5 October 2010 – Published: 6 January 2011

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Published by Copernicus Publications on behalf of the European Geosciences Union.

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## Abstract

The evolution of aerosols in the tropical upper troposphere/lower stratosphere between June 2006 and October 2009 is examined using the observations of the space borne CALIOP lidar aboard the CALIPSO satellite. Superimposed on several volcanic plumes and soot from an extreme biomass-burning event in 2009, the measurements reveal the existence of fast cleansing episodes of the lower stratosphere to altitudes as high as 20 km. The cleansing of the full 14–20 km layer takes place within 1–4 months. Its coincidence with the maximum of convective activity in the southern tropics, suggests that the cleansing is the result of a large number of overshooting towers, injecting aerosol-poor tropospheric air into the lower stratosphere. The enhancements of aerosols at the tropopause level during the NH summer may be due to the same transport process but associated with intense sources of aerosols at the surface. Since, the tropospheric air flux derived from CALIOP observations during North Hemisphere winter is 5–20 times larger than the slow ascent by radiative heating usually assumed, the observations suggest that convective overshooting is a major contributor to troposphere-to-stratosphere transport with concomitant implications to the Tropical Tropopause Layer top height, chemistry and thermal structure.

## 1 Introduction

All natural and human-made short- and long-lived trace gases controlling stratospheric chemistry and radiative balance are emitted at the surface and rapidly lifted up in the tropics by convective systems. They reach the maximum outflow of convective systems near 14 km within a few hours (Gettelman et al., 2002), from where they are transported across the Tropical Tropopause Layer (TTL) into the stratosphere and distributed globally by the Brewer-Dobson circulation (Holton, 1995). Two mechanisms are in competition for the transport across the TTL: a slow ascent within 6–9 months to 20 km by radiative heating of air masses (Holton, 1995; Corti et al., 2005; Yang

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et al., 2008; Fueglistaler et al., 2008) or fast convective overshooting above storms as proposed by Danielsen (1981, 1993). Even if this view is commonly recognized, the monsoon circulations over Asia and Africa during the Northern Hemisphere summer offer an alternative pathway for tropospheric air to reach the lower stratosphere bypassing the TTL (Dunkerton, 1995; Gettelman et al., 2004; Randel et al., 2010).

Due to the time required to ascend through the TTL by slow radiative ascent, very short-lived chemically active species, VSLS, are thought to be deactivated and the air is thought to be dehydrated at the cold point tropopause. Although recognized to exist (Danielsen, 1982, 1993), the contribution to vertical transport across the TTL of fast convective overshooting of adiabatically cooled air is generally believed to be too infrequent to be significant (Fueglistaler et al., 2008 and references therein). However, several recent observations adjacent to or above continental tropical convective systems during recent European balloon and high altitude aircraft campaigns, HIBISCUS (Pommereau et al., 2007) and TROCCINOX (Chaboureau et al., 2007) in Brazil, SCOUT-O3 in Northern Australia (Brunner et al., 2009) and SCOUT-AMMA in West Africa (Cairo et al., 2009), have shown that convective overshooting, well captured by mesoscale cloud resolving models (CRM) (Chaboureau et al., 2008; Grosvenor et al., 2008), are more common than previously thought. They can reach altitudes as high as 20 km and thus strongly impact the thermal structure of the TTL (Pommereau and Held, 2007; Cairo et al., 2009).

Although the frequent occurrence of such events over tropical continents is becoming more widely recognized, their contribution to Troposphere-to-Stratosphere Transport at global scale is still debated. There are indications however that it might be more significant than previously thought. For instance, the precipitation radar of the Tropical Rainfall Measurements Mission-Precipitation Radar (TRMM-PR) indicates that at least 1% of storm turrets reach the TTL (Liu and Zipser, 2005), particularly over Africa (Zipser, 2006; Liu and Zipser, 2009).  $\text{CH}_4$  concentration measured by HALOE,  $\text{N}_2\text{O}$  observed by ODIN-SMR and  $\text{N}_2\text{O}$  and CO from AURA-MLS all display maxima over tropical continental areas (Ricaud et al., 2007; Schoeberl et al., 2008). Finally, an

additional indication provided by the high-resolution aerosols profiles of the CALIOP lidar aboard the CALIPSO satellite, reveals an apparent aerosol cleansing of the TTL and lower stratosphere during the Northern Hemisphere winter that could be linked to convective overshooting (Vernier et al., 2009).

Except for infrequent strong volcanic eruptions (McCormick et al., 1995), the source of aerosols in the tropical Upper Troposphere and Lower Stratosphere is poorly understood. In situ particle measurements have shown that the production of Aitken nuclei ( $r < 50 \text{ nm}$ ) is very high in the tropics compared to the mid-latitudes, involving gas-to-particles conversion of  $\text{SO}_2$  and other gas precursors (DMS) (Bock et al., 1995). Recent studies suggest that non-sulfate aerosols also contribute to the total aerosol mass in this region (Ekman et al., 2006; Bormann et al., 2009). Particle composition and volatility analysis have indicated the presence of aerosols, which could be composed of black carbons and other carbonaceous materials (Fryod et al., 2009). The lifetime of upper tropospheric aerosol is however controlled by convection by which they can be removed and precipitated to the ground (i.e., “washout”). Since at least 1% of tropical convective systems reach the TTL (Liu and Zipser, 2005), the process might be an important sink of aerosols in this region (Hamill et al., 1997). The objective of this paper is to further examine the CALIOP clean air events from the beginning of the mission in June 2006 until October 2009, and their possible relation with convection in the tropics. Section 2 provides a description of CALIOP aerosols observations in the UT/LS, followed in Sect. 3 by a comprehensive analysis of the fall-winter-spring 2007–2008, the optimum period, least affected by volcanic/fires events, for studying the vertical and latitudinal propagation of the clean air. The last section is dedicated to explain the origin of the cleansing and the mechanisms of vertical transport in the UT/LS that can be inferred from those events, followed by an estimation of the mass flux between the troposphere and stratosphere at global scale corresponding to the observed cleansing.

## 2 Cleansing of the UT/LS as seen by CALIOP

The CALIPSO mission, a collaboration between the National Aeronautics and Space Administration (NASA) and the Centre National d'Etudes Spatiales (CNES), is dedicated to the study of clouds and aerosols from the troposphere to the stratosphere (Winker et al., 2007). Part of the A-Train constellation, the satellite is in polar orbit at 705 km, providing measurements at 01:30 and 13:30 LT with a repeat cycle of 16 days. Since June 2006, the CALIOP lidar measures backscatter profiles at 532 nm and 1064 nm, and depolarization at 532 nm with a vertical resolution of around 200 m in the stratosphere (Hostetler et al., 2006). Because of the low signal-to-noise ratio on individual profiles, the detection of aerosols in the stratosphere requires averaging. A method for deriving the Scattering Ratio (SR) (the contribution of aerosol plus molecular backscatter to molecular only) has been developed by Vernier et al. (2009). The procedure is based on the averaging of 532 nm nighttime-only measurements in one degree latitude bands ( $\sim 300$  profiles) and further arranged into a regular grid of  $1^\circ$  latitude  $\times$   $2^\circ$  longitude and 200 m in altitude during the 16-day repeat cycle. To rectify a known CALIOP calibration deficiency that is particularly noteworthy in the UT/LS, all data are corrected using the recalibration technique described in Vernier et al. (2009). Artefacts in the CALIOP SR time series created by sudden changes in the GEOS-5 temperature at 36–39 km, the aerosol free altitude layer used for adjusting the calibration, have been removed by replacing the GEOS-5 temperatures with those of the ECMWF ERA-INTERIM reanalysis found to better match the temperatures observed by MLS/Aura (Schwartz et al., 2008). In addition, a mask for the South Atlantic Anomaly (SAA) has been applied with which all 532 nm profiles showing dark noise greater than 100 photons in this area have been discarded (Hunt et al., 2009). Finally, clouds below 20 km have been filtered by removing all pixels for which the mean volume depolarization ratio is greater than 5%.

Shown in Fig. 1 is the resulting evolution of the CALIOP zonal mean SR between 14–30 km within the tropical band  $20^\circ$  N– $20^\circ$  S (after removing the SAA) from the beginning

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of the mission in June 2006 until October 2009 and in Fig. 2, time/latitude cross sections of the same quantity at four potential temperature levels centred at 380 K (17 km), 420 K (18.5 km), 460 K (20 km) and 500 K (21.5 km). The blank section in February–March 2009 on both figures is due to an interruption of the CALIOP measurements during the laser changeover.

Several volcanic plumes are seen in the tropics (Fig. 1) as well as at mid-latitudes (Fig. 2) displaying SR values greater than 1.2. The dominant one was injected after the volcanic eruption of the Soufriere Hills in Montserrat Island in the Caribbean on 20 May 2006. The plume remains in the tropics at 18–20 km (460 K) for 4–5 months before starting ascending in the tropical stratosphere to reach 25 km within more than a year. The second volcanic feature, less intense, is seen in October–December 2006 after the eruption of the Tavurvur cone of the Rabaul volcano in Papua-New Guinea on 7 October. The lifetime of this plume is drastically reduced compared to the first one, rapidly transported to mid-latitudes at isentropic levels from 360 to 440 K (Fig. 2), and replaced by air of low aerosol concentration in the tropics. Other volcanic plumes are seen during the summers 2008 and 2009 in the Northern Hemisphere (NH) after the eruption of Kasatochi in Alaska (US) on 07 August 2008 and Sarychev in Kamchatka (Russia) on 12 June 2009. Both plumes are further transported to the northern tropics, but more rapidly at lower (380 K) than higher levels (460 K) (Fig. 2), creating an apparent ascent in the tropics (Fig. 1). The remaining feature seen at 480–520 K (21 km) in March–April 2009 after the laser changeover with a SR between 1.10–1.12 is not the signature of a volcano but of soot particles from an extreme biomass-burning episode near Melbourne in Southern Australia on 7 February 2009 (Trepte et al., 2009).

In addition to these events, aerosol layers are also present at 360–400 K from May to November, more intense in the Northern Hemisphere subtropics and at mid-latitudes, when no volcanic events could be identified. The occurrence of the non-volcanic aerosol enhancement occurs every year during this period during the monsoon season, extending from Western Asia to Eastern Mediterranean. The monthly latitude-height cross-sections between June and September in 2007 over Asia (not shown), indicates

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that they don't come from an old volcanic plume transported southward, across the weak barrier afforded by the NH summer jet stream (Chen, 1995; Dunkerton, 1995).

Besides those remarkable features, are the so-called "clean air", of scattering ratio less than 1.04 that start at the tropopause and propagate upward with time. Since the CALIOP lidar at 532 nm is primarily sensitive to particles with radii larger than 30 nm according, a "clean air" event refers to air masses with low densities of such particles. The first event in early 2007 can be seen from 14 to 18 km (360–440 K), more pronounced in the tropics and that rapidly displaced the Tavorvur plume. A similar episode occurred during the same season in 2008, a year undisturbed by volcanoes and fires, the lower stratosphere being cleansed up to 19–20 km and remained relatively aerosol free until the following year between 18 and 21 km (440–520 K).

Though of higher resolution and free of clouds interference in the CALIOP data, the cleansing of the lower stratosphere during the NH winter-spring, resulting in an aerosol annual cycle, was observed by SAGE II at 18 km between 1998–2005 (Thomason et al., 2008). It is also confirmed by independent balloon measurements in the tropics within the SCOUT-O3 project shown in Figure 3 with: i) the aerosol mixing ratio of particles of radius greater than 0.15  $\mu\text{m}$  and 0.25  $\mu\text{m}$  derived from an optical particle counter (OPC, Deshler et al., 2003) during a flight in May 2008 from Teresina, Brazil at 5° S and, ii) the SR measurements of a backscatter sonde (BKS, Rosen and Kjöme, 1991) flown in September 2008 at 12° N in Niamey, Niger. Both are compared to the mean CALIOP SR within a box of  $\pm 7^\circ$  latitude  $\pm 70^\circ$  longitude centred at the respective balloon locations. Both balloon data show a drop of aerosol mixing ratio or backscatter signal between 17 and 20 km compatible with the decrease observed by CALIOP. The peaks below 17 km in the BKS profile are due to the presence of high altitude clouds that are discarded in the CALIOP analysis. The aerosol enhancement observed by CALIOP at and below 17 km in September 2008 is due to the debris from the Alaskan volcano in the northern part of the  $\pm 7^\circ$  latitude box (at 360–400 K in Fig. 2).

Features such as volcanic or soot plumes are observed in the tropical and extra tropical UT/LS during the first 40 months of the CALIPSO mission with unprecedented

details on their vertical and meridional transport. The Soufriere Hills plume is slowly transported in the middle stratosphere at an average velocity of 300 m/months by the Brewer-Dobson circulation consistent with previous estimations (Rosenlof et al., 1997; Mote et al., 1998). The meridional transport of the plume in or out of the tropics is shown to depend on the altitude, faster at lower levels and becoming gradually inhibited at increasing altitude until reaching the tropical pipe (Plumb et al., 1996), consistent with the drop of ozone variability at this level reported from circumnavigating long duration balloon measurements by Borchers et al. (2007). But the volcanic plumes in the TTL and tropical lower stratosphere are shown to disappear more quickly than expected from their meridional transport, influenced by tropical episodes of clean air especially pronounced between 20° S–10° N during the Northern Hemisphere Winter at 14–20 km. The only period undisturbed by additional aerosol input is the 2008 NH winter during which the clean air is observed to propagate rapidly in the TTL and lower stratosphere up to 19–20 km, before being slowly transported by the Brewer Dobson circulation displaying a tape recorder like feature similar to that of water vapor reported by Mote et al. (2008).

### 3 Location and vertical propagation of the cleansing

Since the low signal-to-noise ratio of CALIOP does not allow constructing SR maps between 15 and 20 km with a temporal resolution of less than 2 months, and the Quasi Biennale Oscillation (QBO) was in its easterly phase during the winter 2008 with zonal wind speed of around –15 m/s at 50 hPa (<http://www.cpc.noaa.gov/data/indices/qbo.u50.index>), an air mass in the tropical lower stratosphere was circumnavigating twice during this two-month period, making difficult to determine the longitudinal dependence of the cleansing. As a result, it is more appropriate to look at the bimonthly mean latitude-height cross-sections. Figure 4 shows those two-months cross-sections from October 2007 to September 2008, together with the mean zonal cloud top in red (depolarization ratio >5%), allowing following the evolution of the clean air episodes in more



detail. The cleansing starts in October–November 2007, intensifying in December–January with SR values of less than 1.04 propagating from 14 km to 17 km between 20° S–5° N in phase with the latitude of cloud tops. The cleanest period occurs in February–March where low SR values reach altitudes as high as 19–20 km, 2–3 km higher than the cloud top, making the whole TTL and lower stratosphere virtually free of aerosol. Later in April–May, an isolated bulb of clean air is seen at 18–20 km between 10° N–20° S above cloud top, which stagnates at this level until September 2008. Also seen in April–May is the presence of an aerosol layer at 16–17 km, between 10–30° N, reinforced and extended to higher latitude in June–July, before the arrival of the Kasatochi plume in September. The vertical propagation of the clean air is seen to be disrupted by the arrival of air rich in aerosols from the stratosphere in June–July 2008, consistent with the downward transport of stratospheric air from the tropical reservoir during the easterly phase of the QBO (Treppe et al., 1992), separating the clean air into two upward horns asides from the equator.

To better characterize the relationship between cleansing episodes and convection, we show in Fig. 5 times series of mean SR every 1 km between 14–20 km from June 2006 to February 2009 in the northern and southern tropics, together with the number of Overshooting Precipitation Features (OPF) of the TRMM-PR. The TRMM OPFs are selected by considering mesoscale convective systems of contiguous precipitating area greater than 2000 km<sup>2</sup> with a radar signal greater than 20 dBz reaching at least 14 km (Zipser et al., 2005), with a minimum of 2 flashes as observed from the Lightning Imaging Sensor (LIS) also on TRMM. The flash discrimination is chosen to select only convective systems with strong updrafts inducing hail formation and lightning. All OPFs are extracted with those four conditions from the TRMM database (<http://trmm.chpc.utah.edu/>), from June 2006 to February 2009 and rearranged into bins of 16 days comparable to CALIPSO.

Figure 5 shows that the drop of aerosol at all levels up to 18 km is generally correlated with the high number of overshoots in the southern tropics. In 2006 as well as in 2007, whether in the presence of volcanic aerosols or not, the first signs of cleansing appear

in November (black vertical lines), one month after the rise of the TRMM OPFs in the southern tropics. In 2008, the picture is somewhat perturbed by Kasatochi plume in September. On both years, a maximum cleansing is observed in February up to 17 km, delayed by one month at 18 km and two months at 20 km. Afterward, the aerosol loading increases in coincidence with the rising of TRMM OPFs, but this time in the northern tropics.

## 4 Discussion

### 4.1 Origin of the cleansing

Several mechanisms of cleansing could be evoked for explaining the weak aerosol content of air in the lower stratosphere. One possibility could be aerosol sedimentation, but it is inefficient in this case since particles of 0.1–0.2 µm radius, typically observed in the stratosphere, falling at a rate of 0.1 km/month would required about ~60 months to drop from 20 to 14 km. The second one would be the capture by much larger cirrus cloud particles which themselves sediment. As shown by Sassen et al. (2009), the maximum frequency of cirrus cloud occurrence is observed during the NH winter over tropical continents and Western Pacific. In addition, the freezing-drying mechanism for air parcels entering the very low TTL temperature area and followed by sedimentation of ice crystals formed therein would lead to deplete WV water vapor (WV) as observed (Gettelman et al., 2002). If aerosols served as nuclei for cirrus formation a similar minimum in aerosol would be expected. However, even if this mechanism could be efficient at the cold point tropopause (located according to GPS-COSMIC radio-occultation sounding at around 16.5–17.5 km, Liu et al., 2008), the level of the cirrus cloud top (Fig. 4), another explanation is required to explain the cleansing between 18 and 20 km.

As previously shown, the cleansing of the lower stratosphere is correlated with the southern tropics convective season, but anticorrelated with the Northern Hemisphere

season. In order to understand this apparent discrepancy, we examine in Fig. 6, the position of the TRMM OPFs on bimonthly maps from October 2007 to September 2008, together with the amount of aerosols in the troposphere, given by the total column Aerosol Optical Depth (AOD) from the CALIPSO level 2 profile products (Omar et al., 2009). From December to March, most of the deep convection is observed south of the equator over the Central African, Amazonian and Indonesian tropical forests, in coincidence with low values ( $<0.1$ ) of AODs. In contrast, later in the season, when convection is reaching West Africa in April–May and South-East Asia in June–September, OPFs are associated with significantly higher CALIOP AOD values (up to 0.6).

For investigating the potential transport of aerosol in the TTL, we constructed a TTL Aerosol Index, based on the position of each overshoot (OPF) as seen by TRMM and the surrounding values of AOD from CALIPSO level 2 product. It is given for each month  $t$  by

$$AI(t) = \frac{1}{N} \sum_{n=1, N} OPF(n, t) \times AOD(n, t)$$

where AI is the Aerosol Index,  $n$  the index of each OPF ( $N$  the total) and AOD the value at the OPF location. Figure 7 shows the comparison between this index and the mean CALIOP SR between 14–18 km from October 2007 to September 2008. Low values of AI are observed from October 2007 to February 2008, in coincidence with the beginning of the drop of aerosol in the TTL until the minimum observed in February. Afterward, the AI and SR increase together to reach a maximum during the NH summer 2008.

The relationship between AI and real aerosol loading of the TTL seems to indicate that air masses transported by convection/overshoot from the clean surface over the rainy forests of the southern tropics (Congo, Amazonia, Indonesian Islands) could lead to the low aerosol loading of the TTL during the NH winter. The minimum observed, delayed by 2 months compared to the AI, would be consistent with the progressive cleansing resulting from the multiple overshoot of clean air. On the contrary, the increase of

the aerosol load during NH summer would be accounted for the position of convection, located at the border of desert and polluted areas during the Asian and African monsoon periods, which would be responsible for the transport of particles at higher levels.

## 4.2 Impact of overshooting at the global scale

The mass flux of tropospheric air, injected by overshoot at an altitude  $Z$ , required for explaining the cleansing of the TTL during the SH convective season can be derived from the drop of aerosol during the period November 2007–February 2008 (Fig. 5). Figure 8 illustrates the conceptual model of overshoot that we are using to calculate the time constant of dilution and the total mass flux between the troposphere and the stratosphere related those events. First, we suppose that CALIOP (SR-1) ( $SR-1 = \beta_{aero} / \beta_{mol}$ ) is proportional to an aerosol mixing ratio (assuming that the aerosol phase function is constant). After applying the mass and aerosol mixing ratio flux conservation, we can show (Appendix A1) that the aerosol mixing ratio follows an e-folding law, where  $\tau$ , the time constant required for (SR-1) to decrease by 63%, is directly proportional to the mass flux (Appendix A1).  $\tau$  observed by CALIOP varies from  $1.5 \pm 0.5$  months at 15 km, to  $3 \pm 0.5$  months at 18 km, and  $4 \pm 0.5$  months at 20 km (Table 1). The flux corresponding to the cleansing of each layer at global scale shown in Table 1, is of  $780 \pm 39 \times 10^8 \text{ kg s}^{-1}$  at 15 km and  $240 \pm 60 \times 10^8 \text{ kg s}^{-1}$  at 18 km that is 5 to 12 times higher than values derived from radiative calculations by Corti et al. (2005) and Yang et al. (2008). At higher levels, 18–20 km it is even larger (up 20 times) showing that convective overshooting could be an efficient mechanism to cleanse the lower tropical stratosphere.

## 4.3 Implications for the lower stratosphere

Several other mechanisms could be proposed to account for the rapid vertical propagation of the clean air (Figs. 1 and 2), including the slow ascent by radiative heating, the vertical turbulent diffusivity, and gravity waves. As shown before, the radiative heating

at a rate of less than 0.4 mm/s or 1 km/month (Yang et al., 2008) will take 6–9 months for an air mass to reach 20 km from the tropopause that is too slow to explain the cleansing. The same conclusion could be applied to the average vertical turbulent diffusivity of  $0.02 \text{ m}^2 \text{ s}^{-1}$  derived by Mote et al. (1998) from 5 yr of total hydrogen profiles of the halogen occultation experiment which still required about 5 yr for a one kilometre rise. Even with the upper bound equivalent diffusivity of  $0.5 \text{ m}^2 \text{ s}^{-1}$  derived by Pissu and Legras (2008) from balloon ozone and water vapor profiles of the HIBISCUS campaign in Brazil, thus including turbulent diffusivity, gravity waves and convection, it would take 2.5 months for a one-kilometer apparent uplift. The only remaining option is the frequent injections of clean tropospheric air by convective overshooting that would progressively clean the lower stratosphere. The time constant of dilution, resulting from those injections, would increase with altitude since the overshoot frequency is known to decrease with altitude (Liu and Zipser, 2005). This scheme is consistent with the recent observations of geyser like injection of ice particles in the lower tropical stratosphere over land convective regions (Nielsen et al., 2005; Corti et al., 2008; Khaykin et al., 2009) implying a hydration process of the lower stratosphere, as well as the injection of adiabatically cooled air modifying its thermal structure (Pommereau and Held, 2007; Pommereau et al., 2010). It would also explain the larger  $\text{N}_2\text{O}$ ,  $\text{CH}_4$  and  $\text{CO}$  concentration in the lower stratosphere over tropical continents in contrast to oceans as well as their seasonal variation reported by Ricaud et al. (2007, 2009) or Schoeberl et al. (2008). It will imply a top TTL, the region sharing tropospheric and stratospheric characteristics, around 20 km higher than generally assumed (Fueglistaler et al., 2008). Finally, it will imply also the fast transport of chemically reactive Very Short-Lived Species (VSLS) in the lower stratosphere with their possible impacting on ozone chemistry. However, if the overshooting events and the resulting hydration of the lower stratosphere are well captured by meso-scale cloud resolving simulations (e.g., Chaboureaud et al., 2007; Grosvenor et al., 2007), they are poorly represented by global Chemistry Transport Models (CTM) forced by Numerical Weather Forecast Models (NWP).

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## 5 Conclusions

The three-year CALIOP aerosols profiles in the tropics reveal the existence of a fast cleansing process of the lower stratosphere up to 20 km. It is shown to take place during the Southern Hemisphere convective season, where convective overshooting systems seen by the TRMM precipitation radar are occurring in clean region of the troposphere over large rain forest, consistent with the injection of relatively clean air by overshoots. The progressive dilution of the lower stratosphere by those events could be explained by the repetition of a large number of deep overshooting towers over flown by stratospheric air masses. The tropospheric air flux inferred from the CALIOP observations, 5–20 times larger than the one derived by radiative heating calculation, indicates that convective overshooting is a major contributor of troposphere-to-stratosphere transport during the NH winter with many implications on the TTL top height, hydration, chemistry and thermal structure of the lower stratosphere. Such process, poorly represented by global Chemistry Transport Models (CTM), will be assimilated in the future into an isentropic transport model to study in details the way it could be parameterized in CTM.

## Appendix A

### Overshoot mass flux

As shown by the conceptual model of overshoot in Fig. 8, the mass flux conservation in the TTL can be written:

$$D_c - E_c + D_{\text{ext}} - E_{\text{ext}} = 0 \quad (\text{A1})$$

Where,

–  $D_c$  Is the convective flow ( $D$  for Detrainment) (Unit kg/m/s)

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- $E_c$  is that extracted ( $E$  for Entrainment)
- $D_{ext}$  is the input from horizontal transport and
- $E_{ext}$  the horizontal outflow

5 Neglecting the weak transport from mid-latitudes to the tropics allow simplifying Eq. (A1) by:

$$D_c - E = 0 \quad (\text{with } E = E_c - E_{ext}) \quad (A2)$$

The change of aerosol mixing ratio,  $\chi$ , due to the mixing with clean air can be expressed:

$$\rho \times A \times \frac{\partial \chi}{\partial t} = D_c \times \chi_c - E \times \chi \quad (A3)$$

10 where  $\rho$  is the air density,  $A$  the surface of the layer, and  $\chi_c$  the aerosol mixing ratio in the convective cloud, the solution of Eq. (A3) is:

$$\chi(t) = (\chi_0 - \chi_c) e^{\frac{D_c}{\rho A} \times t} + \chi_c \quad (A4)$$

where  $\chi_0$  is the initial aerosol mixing ratio in the layer and  $\chi(t)$  its change following an e-folding law, with  $\tau = \rho A / D_c$ . We suppose that the aerosol-mixing ratio in the convective clouds is null (clean air,  $\chi_c = 0$ ).

15 Since,  $\tau$  is the time required for reducing the aerosol mixing ratio by 63%, the inflow  $F_c$  within each 1 km thick layer between 15–20 km can be expressed:

$$F_c = D'_c \times \Delta Z = \frac{\Delta Z \rho A}{\tau}$$

20 Assuming that the CALIPSO backscatter ratio ( $SR-1 = \frac{\beta_{mie}}{\beta_{ray}}$ ) is proportional to the aerosol mixing ratio. the tropospheric air flux required for cleansing the TTL by 63%, can be derived from the decrease of CALIOP mean SR from November 2007 to February 2008 between 20° S–0° S shown in Fig. 5 using Eq. (A4). Values are displayed in Table 1.

*Acknowledgements.* This work is part of the PhD thesis of JPV financed by the Centre National la Recherche Scientifique at LATMOS/UVSQ in France now on a NASA Postdoctoral Program at Langley Research Center, administrated by Oak Ridge Associated Universities. The research was supported by the CNES CALIPSO and European SCOUT-O3 projects. The CALIOP data were made available at the ICARE data center (<http://www-icare.univ-lille1.fr/>) and processed at NASA LaRC. The authors want also to thank the University of Utah for providing the TRMM MCS searching system (<http://trmm.chpc.utah.edu/>). The author wants to thank Kristina Ruhlman for making the conceptual view of overshoot in Fig. 7, Jay Kar for the processing of the Aerosol Optical Depth data from the CALIPSO level 2 products and P. Lucker for her help in the processing of the CALIPSO level 1 data.



The publication of this article is financed by CNRS-INSU.

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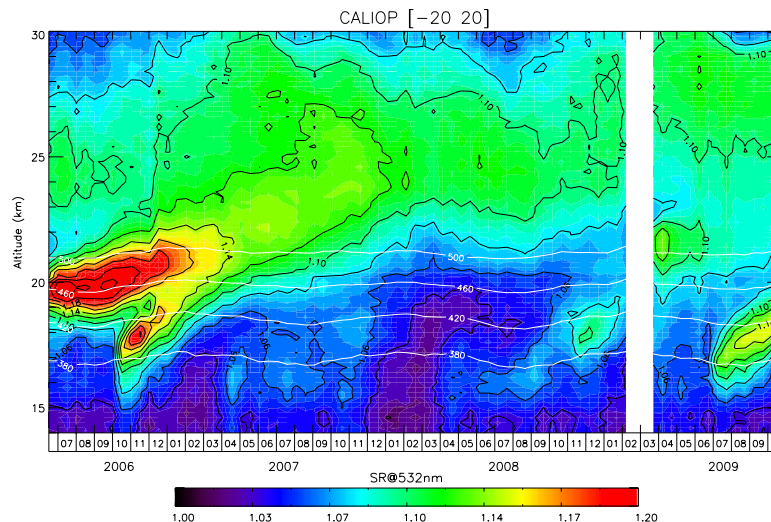
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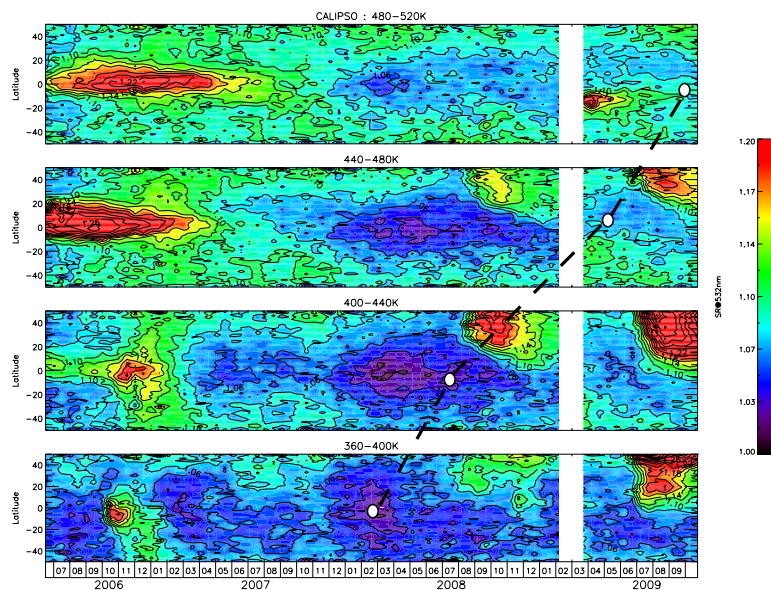
**Table 1.** Average mass flux in  $10^8 \text{ kg s}^{-1}$  and time constant ( $\tau$ ) required for a (SR-1) decrease by 63% in the TTL between  $20^\circ \text{ S}$ – $0^\circ \text{ S}$  from 15 km to 20 km. Values are derived from the dilution model by overshooting (Appendix A1) and compared to radiative heating calculation by Corti et al. (2005) on yearly average basis and by Yang et al. (2008) during the NH winter only.

Altitude	Mass flux from CALIPSO ( $10^8 \text{ kg s}^{-1}$ )	From Corti et al. (2005)	From Yang et al. (2008)
15 km	$780 \pm 39$ ( $\tau = 1.5 \pm 0.5$ months)	139	
16 km	$670 \pm 340$ ( $\tau = 1.5 \pm 0.5$ )	102	56
17 km	$430 \pm 160$ ( $\tau = 2 \pm 0.5$ )	65	74
18 km	$240 \pm 60$ ( $\tau = 3 \pm 0.5$ )	19	28
19 km	$200 \pm 50$ ( $\tau = 3 \pm 0.5$ )	9	9
20 km	$120 \pm 20$ ( $\tau = 4 \pm 0.5$ )	9	9



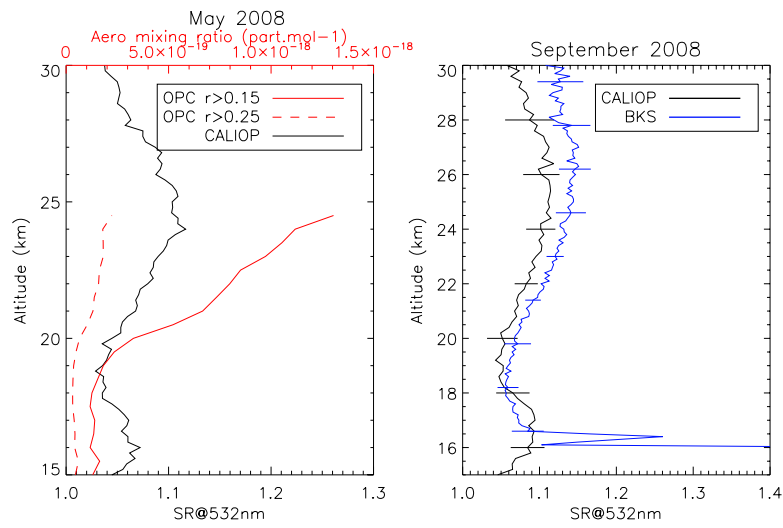
**Fig. 1.** Mean CALIOP Scattering Ratio (SR) (contour steps 0.02) between 14–30 km and 20° S–20° N from June 2006 to October 2009 after removing cloudy pixels with a depolarization ratio at 532 nm greater than 5%. The precision of the SR profiles is 2%, the vertical resolution 200 m and the time resolution 16 days. Shown by white lines are temperature potential levels at 380, 420, 460 and 500 K.

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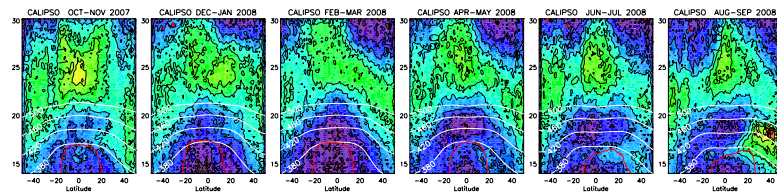


**Fig. 2.** Latitude-time cross-sections of CALIOP SR within 40 K (1.5 km) thick layers centred at 380 K (17 km), 420 K (18.5 km), 460 K (20 km) and 500 K (21.5 km) from June 2006 until October 2009. The black dotted line shows the timing of the uplift of clean air if it was due to slow ascent by radiative heating as calculated by Yang et al. (2008).

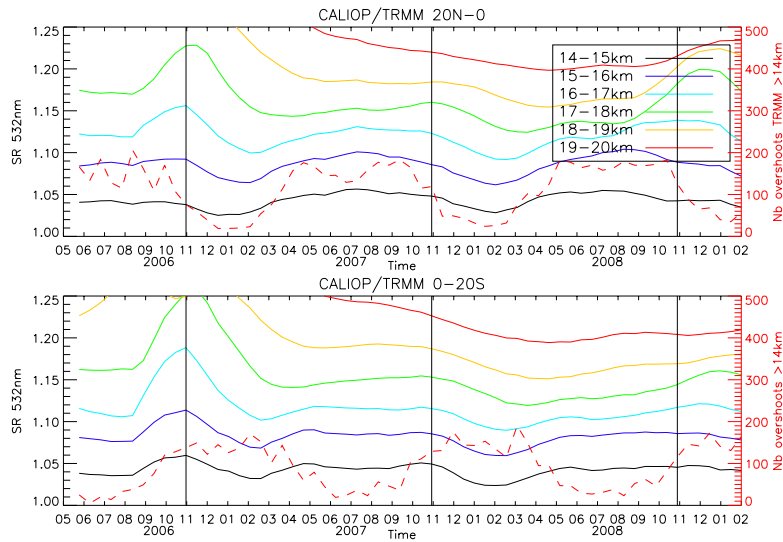
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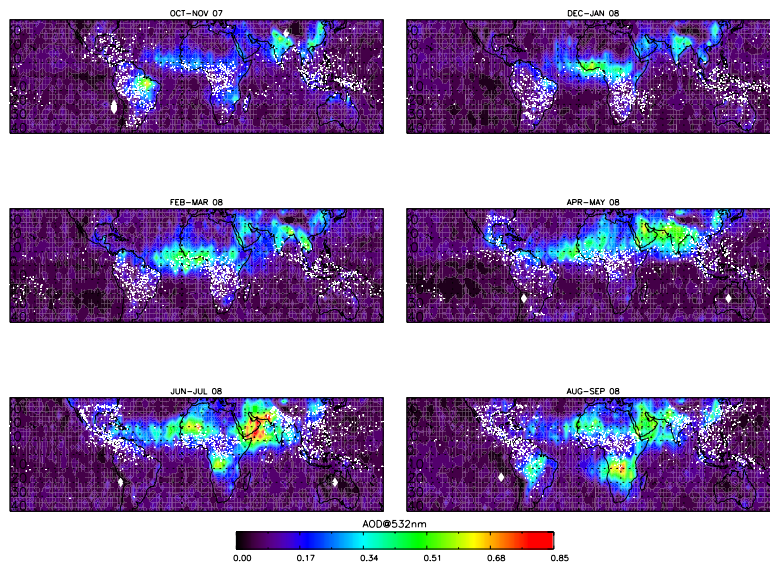
**Fig. 3.** Left: Aerosol mixing ratio of particles of radius  $>0.15 \mu\text{m}$  and  $>0.25 \mu\text{m}$  observed by a balloon-borne Optical Particle Counter at  $5^\circ \text{S}$  in Brazil in May 2008; right: mean scattering ratio reported by a BKS sonde at  $12^\circ \text{N}$  in Niger in September 2008; both compared to CALIOP SR within a box of  $\pm 7^\circ$  latitude  $\pm 70^\circ$  longitude centred at the location of the balloon flight.



**Fig. 4.** Latitude-height cross-sections of mean CALIOP SR from September 2007 to August 2008 every two months. Shown by white lines are temperature potential levels at 380, 420, 460 and 500 K and by red line mean cloud top height (Zonal mean depolarization ratio=5%).

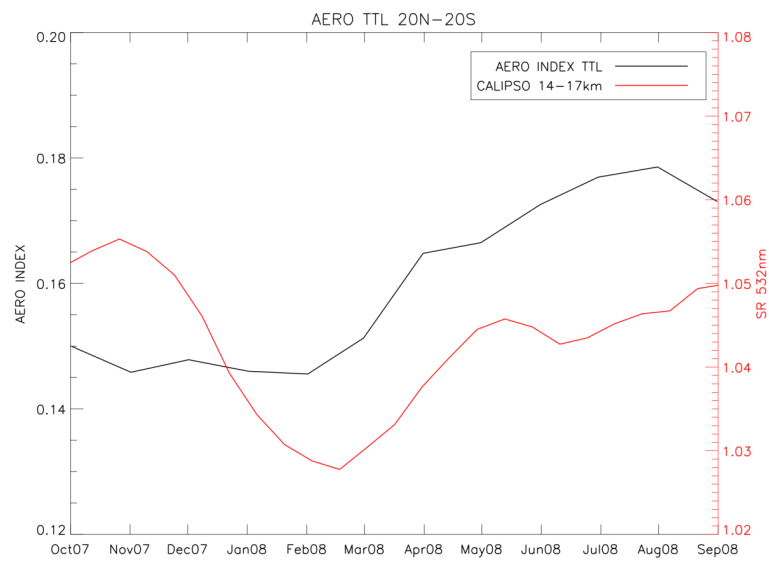


**Fig. 5.** Time series of the zonal mean SR between 0–20° N (upper panel) and 0–20° S (lower panel) every kilometer from 14 km to 20 km separated vertically by an offset of 0.02. The red dotted line represents the number of overshoot reaching an altitude higher than 14 km as seen by the TRMM satellite.

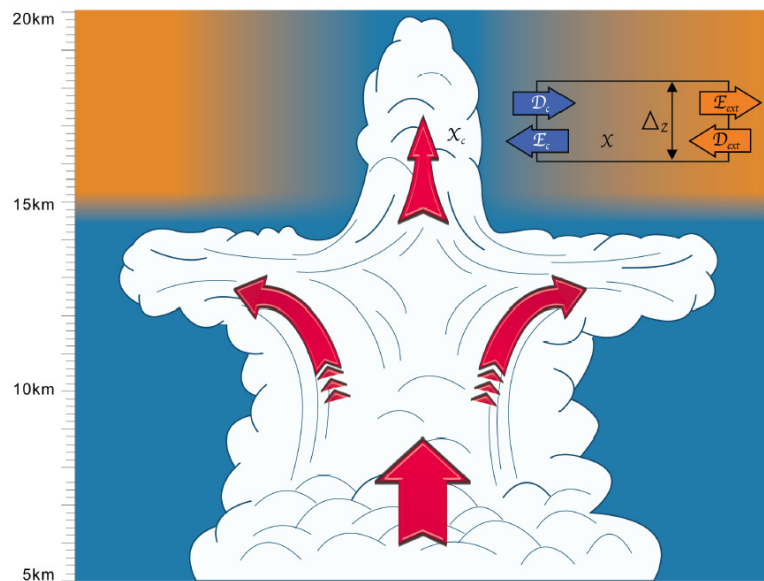


**Fig. 6.** Bi-monthly average of Aerosol Optical Depth (Total column) from CALIPSO level 2 products from October 2007 to September 2008 and location of TRMM Overshoot Precipitation Features (OPF) over the same period (white dots).





**Fig. 7.** Mean SR between 14–17 km and TTL Aerosol Index derived from TRMM OPF and CALIPSO AOD from October 2007 to September 2008.



**Fig. 8.** Schematic of the TTL aerosol-cleansing model by overshooting towers.