

It is now widely accepted that most of the transport of tropospheric air into the stratosphere takes place in the tropics across what is now known as the Tropical Tropopause Layer (TTL). This layer, whose vertical extent is over several kilometres, spans the gradual transition from a region dominated by the convectively overturning circulation of the Hadley cell to a region of slow ascent, controlled by the wave-driven tropical branch of the Brewer-Dobson circulation. The chemical and physical processes that take place within the TTL can strongly influence the chemical distribution of the stratosphere by e.g. providing the necessary catalysts for ozone destruction (mainly composed of the very short lived (VSLS) substances), water vapour and long-lived pollutants like airplane exhausts. The TTL thus plays a vital role in regulating global climate. It therefore follows that understanding the processes that control the distribution of tracers within the TTL is important not only from a scientific point of view but also from an environmental point of view as it allows to plan environmental policies on a firmer ground.

The processes that affect the distribution of tracers within the TTL are very complex, and operate on a large range of scales: from micro-physical and chemical processes to mixing induced by medium-scale turbulent convection, and planetary-scale transport. The interplay of these processes result in highly variable tracer distributions. Their evolution remains particularly challenging for water vapour and VSLS.

The contribution of this project can be divided into two parts: the first part uses an idealized approach in order to understand the role of mixing on the distribution of water vapour. The strongly non-linear character of condensation means that it cannot be simulated accurately using low-order numerical schemes. We therefore developed a high-order numerical code in order to capture the evolution of fine-scale structures in the distributions. We find that the effect of mixing among air parcels does not significantly alter the distributions as long as the sources of moisture and the locations where condensation takes place are sufficiently far apart. This situation is particularly relevant in the subtropics. However if these are close together, the effect of mixing becomes important. The importance of mixing depends on relative magnitude of the mixing time to the transport time between the source and the location of where condensation takes place. We use optimization theory to deduce this result for a passive, non-condensing tracer. The corresponding result for a condensing tracer remains a challenge.

The second part employs an interdisciplinary approach to determine the spatial and temporal distribution of deep convective sources. To that end, we use a novel method that combines a trajectory approach with high-resolution fields of cloud top heights, where the latter are estimated using satellite brightness temperature measurements, obtained from the CLOUD Archive User Service (CLAUS). Unlike previous methods, the time history of air parcels terminates at their crossing of a convective anvil. This way, we do not need to consider unresolved convective motions that would otherwise need to be parameterized. Using this method we were able to thoroughly examine the relative importance of convective sources and find that these are not only highly-localised (see Fig. 1) but also that a small set of them has a long-lasting effect on the upper parts of the TTL. Initially, this effect is localized (see Fig. 2). However most air parcels originating from these sources experience strong horizontal mixing as they rise towards higher altitudes. As a result, a season later, the effect of the

sources is apparent within the whole tropics while beyond a season, higher latitudes are affected. An important result we find is that the radiative effect of clouds is crucial not only for modifying the spatial and temporal distribution of convective sources but also for accelerating the transport of air parcels within the TTL. This result has important implications on the distributions of VSLs within the TTL whose short lifetimes could limit their presence at higher altitudes. With the aid of clouds, their presence could be more important than we previously thought.

For the future and on the basis of this Marie-Curie project, I aim to firstly further my understanding of mixing on the distribution of condensing tracers and secondly to possibly apply the method for determining convective sources to a General Circulation Model in order to examine how well is convection represented in it.