A modelling study of tropospheric distributions of the trace gases CFCl₃ and CH₃CCl₃ in the 1980s

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Abstract. Interhemispheric transport is a key process affecting the accuracy of source quantification for species such as methane by inverse modelling, and is a source of difference among global three-dimensional chemistry transport models (CTMs). Here we use long-term observations of the atmospheric concentration of long-lived species such as CH₂CCl₃ and CFCl₃ for testing three-dimensional chemistry transport models (CTMs); notably their ability to model the interhemispheric transport, distribution, trend, and variability of trace gases in the troposphere. The very striking contrast between the inhomogeneous source distribution and the nearly homogeneous trend, observed in the global ALE/GAGE experiments for both CH₂CCl₃ and CFCl₃ illustrates an efficient interhemispheric transport of atmospherically long-lived chemical species. Analysis of the modelling data at two tropical stations, Barbados (13°N, 59°W) and Samoa (14°S, 124°W), show the close relationship between inter-hemispheric transport and cross-equator Hadley circulations. We found that cross-equator Hadley circulations play a key role in producing the globally homogeneous observed trends. Chemically, the most rapid interaction between CH₂CCl₃ and OH occurs in the northern summer troposphere; while the most rapid photolysis of CH₂CCl₃ and CFCl₃, and the chemical reactions between CFCl₃ and O(¹D), take place in the southern summer stratosphere. Therefore, the cross-equator Hadley circulation plays a key role which regulates the southward flux of chemical species. The regulation by the Hadley circulations hence determines the amount of air to be processed by OH, O(¹D), and ultraviolet photolysis, in both hemispheres. In summary, the dynamic regulation of the Hadley circulations, and the chemical processing (which crucially depends on the concentration of OH, O(¹D), and on the intensity of solar insolation) of the air contribute to the seasonal variability and homogeneous growth rate of observed CH₂CCl₃ and CFCl₃.

Key words: Atmospheric composition and structure (middle atmosphere – composition and chemistry; pollution – urban and regional) – Meteorology and atmospheric dynamics (convective processes)

1 Introduction

The atmospheric transport of long-lived chemical species by large-scale dynamics is the most important process in the redistribution of trace gases in the atmosphere. The long-range transport of pollutants away from a source area, by the large-scale general circulations provides a key mechanism for non-indigenous pollution events. Hence, it is crucial to study the performance of global three-dimensional (3-D) CTMs with respect to their ability to model transport processes. One way of testing CTM’s performance is by studying its ability to model long-lived trace gases, where emission sources are well known and long-term measurements exist, such as chlorofluorocarbons (CFCs).

Trace gases of anthropogenic origin, which contain chlorine and bromine atoms can greatly perturb stratospheric ozone concentrations (e.g. see Solomon, 1999). Most of these man-made chemical species are long-lived. For example, the inferred lifetime for CFCl₃ is about 43–50 years and 4.1–5.4 years for CH₂CCl₃ (Kaye et al., 1994). The longevity of these halogenated species greatly enhances the probability and magnitude of their stratospheric influx. Once they reach the middle atmosphere, they will be destroyed by ultraviolet photolysis and by chemical reaction with O(¹D), releasing active chlorine and bromine atoms into the stratosphere. The subsequent threat to ozone can be examined by detailed examination of catalytic chemistry involving chlorine or bromine-containing compounds (e.g. Wayne, 1993).

The ALE/GAGE experiment, which began in 1978, provides a continuous long-term record of surface observations of the concentrations of the atmospheri-
cally long-lived chemical species (Prinn et al., 1983, 1992; Cunnold et al., 1994). The measurements include two gases which have both anthropogenic and biogenic sources (CH$_4$ and N$_2$O) and five gases of anthropogenic origin (CFC$_1_1$, CF$_2$Cl$_2$, CF$_2$CIFCl$_2$, CH$_3$CCl$_3$, and CCl$_4$). The use of this long-term surface observational record of CH$_3$CCl$_3$ and CFC$_1_1$ from global ALE/GAGE network stations, combined with their anthropogenic emissions, which are reasonably well known and the well-established chemistry (CFCs, HFCs, and HCFCs) by laboratory studies, provide a sound platform for a modelling study. The purpose of this modelling study can be summarized by the following: to derive the three-dimensional distributions of CFC$_1_1$ and CH$_3$CCl$_3$; to study the atmospheric transport processes once these species have been released into the air; and to investigate their trend and variability.

If we consider the following two facts: first there is a near globally homogeneous trend in the concentration of CH$_3$CCl$_3$ and CFC$_1_1$ observed during the ALE/GAGE experiments; second, nearly 95% of the reported sources of those two species are in the Northern Hemisphere with maxima centred around mid-latitudes. Then, the very striking contrast between inhomogeneous source distribution and nearly homogeneous observed trend in the global ALE/GAGE stations, strongly suggests an efficient inter-hemispheric transport of anthropogenic chemical species. This indication of efficient inter-hemispheric transport also emerged from another modelling study of $^{85}$Kr. (Müller and Brasseur, 1995).

The variability of the observed surface concentration of chemical species by atmospheric transport processes has been pointed out by other studies (Prather et al., 1987; Prinn et al., 1992; Hartley et al., 1993). The role of vertical transport by cloud convection on the observed seasonal cycle of the surface concentration has been noted by the modelling study of CFC$_1_1$ and CF$_2$Cl$_2$ over the continental areas (Prather et al., 1987). Cloud convection also plays an important role with respect to interhemispheric transport of trace gases (Gilliland and Hartley, 1998). The variation of the northern Atlantic storm tracks has a direct influence on the observations at Mace Head in Ireland (Hartley et al., 1993), and the ENSO event could have a strong influence of the influx of Northern Hemispheric air in the boreal winter (Prinn et al., 1992). Hence a close relationship between specific atmospheric transport process and the observed variability of surface concentration has long been inferred. The focus of this modelling study is to specifically investigate the observed variability of surface concentration by a variety of atmospheric transport processes. We will concentrate our analysis on the process of inter-hemispheric transport, because it is the key factor for the variability and globally homogeneous trends of concentration of chemical species.

2 Model description

The 3-D CTM (Stockwell and Chipperfield, 1999) used in this study includes parametrizations for transport by subgrid scale processes and chemical reactions which lead to the destruction of CH$_3$CCl$_3$ and CFC$_1_1$. The parametrizations of sub-grid scale transport in the model include cumulus convection, atmospheric boundary layer processes and sub-grid scale diffusive transport.

The cumulus parametrization method (Tiedtke, 1989) used in this study is a mass flux scheme (Stockwell and Chipperfield, 1996). The Tiedtke (1989) cumulus scheme considers the cloud model with the following components: in-cloud updraught, downdraught, large-scale subsidence induced by cumulus motion, entrainment and detrainment processes into in-cloud updraught and downdraught air. The scheme therefore considers only the cumulus scale updrafts, as well as downdraughts, but neglects mesoscale circulations. The closure of the scheme is based on the observational evidence that typical cloud ensembles prevail under certain synoptic conditions. For example, the pre-existence of large-scale low level convergence could moisten and destabilize the environment at low levels, so that small-scale thermals can easily reach the level of free convection and produce deep cumulus convection (Holton, 1992). The key point for the whole mechanism to work is that the low-level large-scale convergence produces a convergence of moisture, which by increasing the equivalent potential temperature in the boundary layer makes the environment more favorable for the development of the cumulus convection.

The transport within the atmospheric boundary layer (ABL) has been parametrized by the local-K scheme (Louis, 1979; Hemmann, 1994; Holtslag and Boville, 1993) plus a simplified boundary layer height calculation to enhance vertical transport of chemical species by large eddy transport in the atmospheric boundary layer. The assumption of the calculation of boundary layer height used in this study is based on the observations of the diurnal variation of boundary layer height (Driedonks, 1982; Deardorff, 1972). The lowest boundary layer height is assumed to be 1 km, and a sine wave variation with amplitude of 2 km is added according to the time of the day. The maximum boundary layer height is therefore 3 km at noon with a minimum of 1 km at midnight. This amplitude is comparable with that calculated by the nonlocal scheme (Holtslag and Boville, 1993) where the diurnal amplitude ranges from 0.5 km to 4 km according to different geophysical locations. The purpose of the boundary layer height is to give an estimated vertical scale of dry convection, which is then used to provide an additional mechanism for vertical transport of species within the ABL. The effect of horizontal diffusive mixing induced by the cumulus convection has been applied to the cloudy model column to account for the enhanced sub-grid scale horizontal entrainment and detrainment of air into the cloud (Stockwell and Chipperfield, 1999), which it is not possible to properly represent in the low-resolution model, and which was shown to affect the interhemispheric tracer transport over the intertropical convergence zone (Prather et al., 1987).

A simple gas phase chemistry scheme, includes the reaction between CH$_3$CCl$_3$ and OH, and CFC$_1_1$ with