LETTERS
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Age of stratospheric air unchanged within uncertainties over the past 30 years

A. Engel[1*], T. Möbius[1], H. Bönisch[1], U. Schmidt[1], R. Heinz[2], I. Levin[2], E. Atlas[3], S. Aoki[4], T. Nakazawa[4], S. Sugawara[5], F. Moore[6†], D. Hurst[6†], J. Elkins[6], S. Schauffler[7], A. Andrews[6] and K. Boering[8]

The rising abundances of greenhouse gases in the atmosphere is associated with an increase in radiative forcing that leads to warming of the troposphere, the lower portion of the Earth's atmosphere, and cooling of the stratosphere above[1]. A secondary effect of increasing levels of greenhouse gases is a possible change in the stratospheric circulation[2,3], which could significantly affect chlorofluorocarbon lifetimes[4], ozone levels[5,6] and the climate system more generally[7]. Model simulations have shown that the mean age of stratospheric air[8] is a good indicator of the strength of the residual circulation[9], and that this mean age is expected to decrease with rising levels of greenhouse gases in the atmosphere[10]. Here we use balloon-borne measurements of stratospheric trace gases over the past 30 years to derive the mean age of air from sulphur hexafluoride (SF_6) and CO_2 mixing ratios. In contrast to the models, these observations do not show a decrease in mean age with time. If models are to make valid predictions of future stratospheric ozone levels, and of the coupling between ozone and climate change, a correct description of stratospheric transport and possible changes in the transport pathways are necessary.

An increase in the rate of upwelling in the tropical lower stratosphere is predicted by all atmospheric general circulation models[2] and is consistent with the observed long-term temperature decrease in the tropical tropopause region[11]. If this increased upwelling also leads to an increase in the overall residual circulation of the stratosphere (Brewer–Dobson circulation), then shorter transport times and shorter residence times of some greenhouse gases and ozone-depleting substances are expected, as these are mainly photolysed at altitudes above 20 km in the tropics, leading to decreased atmospheric lifetimes. An increased upwelling that is restricted to the lowest part of the tropical stratosphere, however, would have a much smaller feedback on the lifetimes of ozone-depleting substances.

The mean age of air is the average time it takes to transport an air parcel from its entry point in the stratosphere, the tropical tropopause, to a given location in the stratosphere. Mean age can be determined empirically from measurements of very long-lived trace gases such as sulphur hexafluoride (SF_6) and CO_2, which increase with time in the troposphere and have neither atmospheric sinks nor sources of significance in the relevant parts of the atmosphere. Stratospheric observations of the two mean-age tracers SF_6 and CO_2 have been compiled from 27 high-altitude balloon flights[12–19] (up to 35 km altitude, SF_6 data from one flight are available up to 43 km altitude) from 1975 to 2005 at Northern Hemisphere mid-latitudes between 32° N and 51° N. Many whole air samples from these flights are archived and have recently been re-analysed for SF_6, and other measurements were made *in situ* during the balloon flights. We were careful to use only high-quality observations with sufficient altitude coverage that are directly linkable to the tropospheric reference time series (see Supplementary Information). For CO_2, data of sufficient quality are available for the period from 1986 to 2005. For SF_6, the selected data span the years from 1975 to 2005, with a gap between the years 1985 and 1994. Note that most profile observations are from the May to October period, when stratospheric variability in the Northern Hemisphere is expected to be lower than during the winter period.

On the basis of the measured time series of tropospheric and stratospheric mixing ratios of these two gases, it is possible to calculate the mean age of mid-latitude stratospheric air masses back to 1975. Figures 1 and 2 show the vertical profiles of CO_2 and the derived mean-age profiles. In general, the mean age increases with altitude up to about 24 km and remains largely constant above that altitude. These vertical mean-age profiles reflect the transport pathways in the stratosphere. Although the variations below 24 km are interesting to study, the data sets available are insufficient to remove the atmospheric variability in this lower altitude region for years before 1992. For this reason, the roughly constant mean-age region above 24 km is much better suited for an investigation of long-term changes of the stratospheric mean age[12]. As altitude is calculated from the pressure measurements on-board the balloon gondolas, we use a criterion of a maximum pressure of 30 hPa (ref. 12), rather than a minimum altitude of 24 km to filter the observations. All data for pressure altitudes between this reference level (shown in Fig. 2 as red line) and 35 km altitude are averaged for each balloon flight to get a representative value of stratospheric mean age for mid-latitudes at that time (see Supplementary Information, Table S1).

Using the data shown in Fig. 2, as well as mean ages derived from vertical profiles of SF_6 mixing ratios, which were treated in the same way as CO_2 values to derive the respective mean ages, we have investigated the long-term evolution of the mean age of stratospheric air. The average mean ages derived from both SF_6 and CO_2 observations are plotted versus the year of sampling in

[1]Institute for Atmospheric and Environmental Sciences, Goethe Universität Frankfurt, 60483 Frankfurt, Germany, [2]Institut für Umweltphysik, Universität Heidelberg, 69120 Heidelberg, Germany, [3]Division of Marine and Atmospheric Chemistry, University of Miami, Miami, Florida 33149, USA, [4]Centre for Atmospheric and Oceanic Studies, Tohoku University, Sendai 980-8578, Japan, [5]Miyagi University of Education, Sendai 980-0845, Japan, [6]Earth System Research Laboratory, NOAA, Boulder, Colorado 80305, USA, [7]Atmospheric Chemistry Division, NCAR, Boulder, Colorado 80307-3000, USA, [8]Departments of Chemistry and of Earth and Planetary Science, University of California, Berkeley, California 94720-1460, USA. †Also at Cooperative Institute for Research in Environmental Sciences, University of Colorado, Boulder, Colorado 80309, USA. *e-mail: an.engel@iau.uni-frankfurt.de.

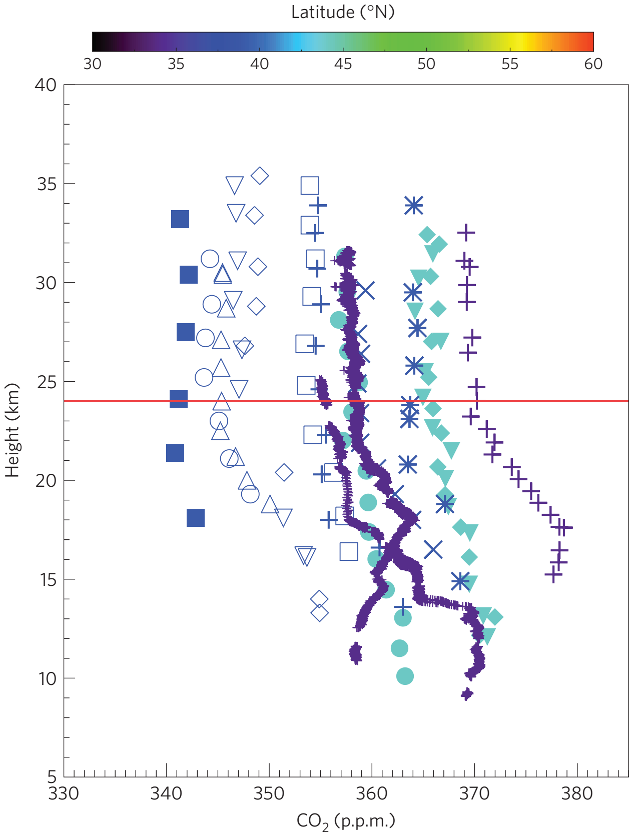

Figure 1 | Vertical profiles of CO_2 in the mid-latitude stratosphere. Data are from measurements of whole air samples collected cryogenically from balloons or from *in situ* measurements on-board a balloon gondola. The data cover the time period between 1985 and 2005. Only data for which a clear link exists to the World Meteorological Organization calibration scales used for this study are included. The colour code shows the (northern) latitude of the measurements. The red line shows the 24 km level, which corresponds to the 30 hPa level chosen as the lower pressure altitude for data included in this analysis. The analytical uncertainty is smaller than the size of the symbols.

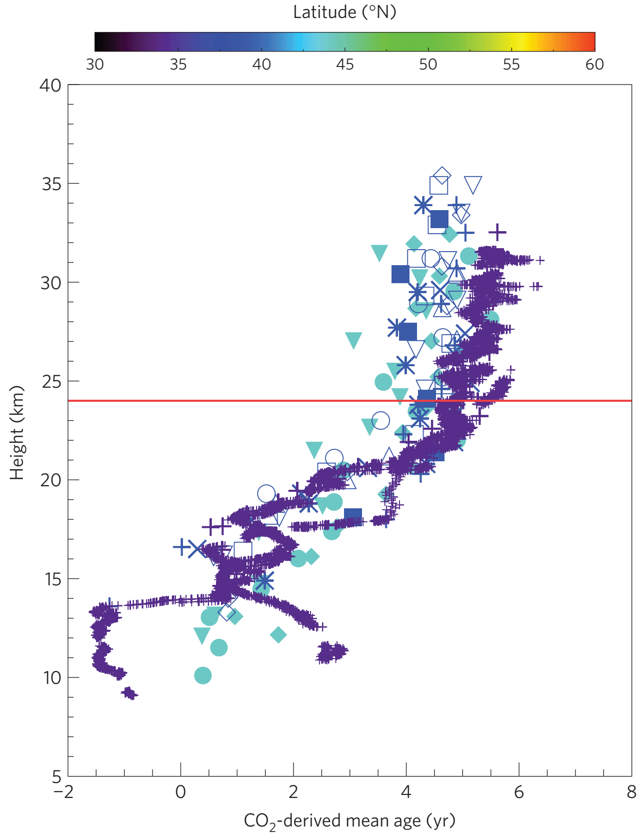

Figure 2 | Vertical profiles of mean age derived from the CO_2 data shown in Fig. 3. The mean age is derived in the same way from the CO_2 observations, as explained in Engel *et al.*[20] using the reference tropospheric data set as discussed in the text. The colour code shows the (northern) latitude of the measurements. The red line shows the 24 km level, which corresponds to the 30 hPa level chosen as the lower pressure altitude limit of data included in this analysis. The uncertainty due to analytical error is of the order of 0.1 years. Systematic uncertainties are discussed in Supplementary Information.

Fig. 3. Although both age tracers usually yield similar results for mid-latitudes[20], the CO_2-derived mean-age values from the years 2001 and 2002 seem to be systematically lower than those from other years and are also lower than the mean-age values derived from SF_6 for the same samples, where available. As there is no reason to exclude these data (see Supplementary Information), we have chosen to include them in the further analysis and to combine the information from both age tracers for the investigation of possible long-term trends. A linear fit to the data, taking into account both systematic and statistical uncertainties (see the Methods section) yields a slope of $+0.24 \pm 0.22$ years of mean age per decade with a mean value of mean age of 4.9 ± 0.5 years. The 90% confidence interval of the long-term evolution derived from our observations is between +0.6 and −0.12 years per decade. We have tested the robustness of this trend by undersampling the input data set. When removing up to six flights arbitrarily from the data set (more than 20%), the mean slope remains at 0.24 years of mean age per decade, with a 1 sigma variability of 0.04 years per decade. The smallest slope calculated after randomly removing six flights is +0.08 years of mean age per decade. In addition, we have repeated the calculation by systematically excluding each of the data sets used for the trend analysis. Under these circumstances, the smallest trend was +0.2 years per decade (when removing the LACE data), and the largest trend was +0.35 years per decade (when removing the NCAR flights dating back to 1975). No negative slope was calculated under any of these sensitivity tests and all of the calculated slopes are well within the range of the estimated overall uncertainty. On the basis of these sensitivity tests and the slope of $+0.24 \pm 0.22$ years per decade, an overall change in mean age of $+0.72 \pm 0.66$ years (1 sigma) is thus estimated for the 30-year time period between 1975 and 2005. On the basis of our error estimate, we conclude that the mean age of stratospheric air above 24 km has shown a significant long-term trend at the 68% confidence level (1 sigma), but at the 90% confidence level the trend is not statistically different from zero. This is consistent with previous investigations on long-term evolution of mean age, using substantially shorter data sets, which were unable to detect significant trends in mean age[12,15]. The long-term trend of 0.72 years over the past 30 years is also of the same order of magnitude as the observed variability of the mean ages determined here (0.5 years), leading us to the conclusion that the observed variability is dominated by natural variations in atmospheric transport that produce variability in tracer mixing ratios in the stratosphere[21].

One model simulation[10,22] suggests a negative trend of stratospheric mean age of up to 20% for the time period between 1960 and 2000, with no significant trend between 1960 and 1975. This result is not consistent with our observation-based trend for the time period 1975 to 2005 at the 95% confidence level. Another model predicts significantly smaller decreases in mean age[23,24], which are

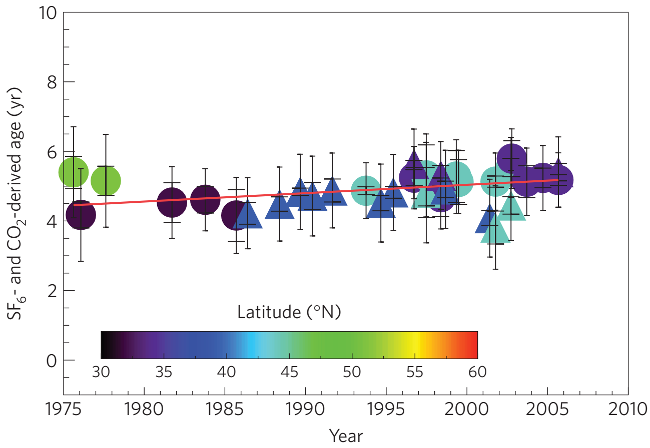

Figure 3 | Long-term evolution of mean age above 24 km altitude. SF_6-derived age values are shown by circles; triangles denote CO_2-derived mean age. The colour code shows the (northern) latitude of the measurements. The inner error bars show the 1 sigma standard deviation of the mean-age values between 24 and 35 km for each individual flight. The outer (larger) error bars denote the overall uncertainty of the mean-age value, including an assessment of the representativeness of a single profile observation (see the Methods section and Supplementary Information for more details). The overall uncertainty has been taken into account for the calculation of the long-term trend.

of the order of −0.25 years over the past three decades, that is, about −0.08 years per decade. Such a decrease in mean age is inconsistent with our observations on the 68% (1 sigma) confidence level but falls within our 90% confidence interval.

We emphasize that our observations do not exclude an increased upward mass flux in the tropics. However, it seems that any increase in this upward mass flux does not result in decreased age of air in the northern mid-latitude stratosphere between 24 and 35 km. If indeed the upward mass flux into the stratosphere has increased, as indicated by other observations in the tropics and predicted by many models, then either other stratospheric transport patterns have also changed[9], masking this effect, or the extra air transported into the stratosphere is not transported to the middle stratosphere for which our analysis is valid. This would imply that the upward mass flux through the tropical tropopause is accompanied by increased poleward transport in the lower stratosphere below 24 km, the lower boundary of our study. This would be in agreement with model calculations suggesting that the trend in the upward mass flux is highest in the lowest part of the stratosphere[3]. In the lower stratosphere, the meridional transport of air masses from the tropics to the mid-latitudes is very effective[25], whereas it is much slower in the middle stratosphere. This results in a transport barrier, often referred to as the tropical pipe[26], which isolates the tropical stratospheric reservoir[27] from mid-latitudes.

In summary, we conclude that this extensive record of stratospheric observations of long-lived tracers shows variability that is in part due to natural variability, as caused, for example, by inter-annual variations in transport. However, at the 90% confidence level, the data show no significant long-term trend in mean age in the Northern Hemisphere mid-latitudes between 24 and 35 km altitude. Rather the observations of both CO_2 and SF_6 as mean-age tracers indicate that the mean age in this region of the stratosphere has remained constant (4.9 ± 0.5 years) over the past three decades. If tropical upwelling has indeed increased during this time, we suggest that this may have resulted in an enhanced circulation in the lower stratosphere, that is, in the so-called tropically controlled transition layer[25]. In the light of the non-significant trend in mean age above 24 km presented here, we suggest that a systematic investigation of possible changes in dynamical tracers in the lower to middle stratosphere at middle and high latitudes may provide the relevant information to explain why increased tropical upwelling has not decreased mean ages above 24 km altitude in the mid-latitude stratosphere. Furthermore, there is also a possibility that trends in mean age over the period 1975 to the present may have been overestimated in some models, owing to the feedback of a high ozone-depletion bias[10,22] and the Brewer–Dobson circulation. Overall, if models are to make valid predictions of future stratospheric ozone levels, and of the coupling between ozone and climate change, a correct description of stratospheric transport and possible changes in the transport pathways are necessary. The comparison of model-derived mean age with the information obtained from tracer observations is a critical test for the model's ability to reproduce stratospheric transport. This data set and analysis provides a strong constraint on the range of plausible trends predicted by models. More stringent constraints on model predictions of long-term change in mean ages of stratospheric air masses will require longer time series of data.

Methods

The distribution of all possible transit times from the tropical tropopause to a certain location in the stratosphere cannot be measured directly. Its mean value (mean age) however, can be derived from the measurements of long-lived tracers, using assumptions about the shape of the age spectrum and also about the temporal development of the trace gas of interest at the entry point to the stratosphere. In this section, we will describe the method used to derive mean age and discuss possible uncertainties, including the representativeness of our balloon observations, which are only brief snapshots of the stratospheric trace-gas composition.

To determine the mean age of air, the width Δ of the age spectrum is parameterized as a function of the mean age Γ, ($\Delta^2/\Gamma = 0.7$), as suggested by Hall and Plumb[28]. A second-order polynomial is fitted to the tropospheric data averaged between 20° N and 20° S for the respective age tracer using variable fit intervals (shorter intervals for younger ages and longer intervals for older ages). For more details, see Supplementary Information and Engel *et al.*[20].

The uncertainty of the derived mean-age values depends on the precision and accuracy of the underlying atmospheric observations, which can be easily assessed. However, it also depends on the accuracy of the assumptions made in the calculation of mean age, for example regarding the temporal evolution of the input of the trace gases of interest into the stratosphere and how well this evolution can be described by the second-order polynomial used in our method. Furthermore, the balloon measurements present snapshot observations that are influenced by shorter-term atmospheric variability, which needs to be distinguished from possible long-term trends. The sensitivity of the mean age to these uncertainties has been quantified by varying the different input parameters within the expected uncertainties. For the mean-age determination, the stratospheric observations must be compared with tropospheric reference time series. Uncertainties in these reference time series will lead to uncertainties in mean age. If the input region to the stratosphere varies with time, this would lead to a change in mean age with time. If this uncertainty in the characterization of the input function remains constant with time (for example, if air is transported preferentially from one hemisphere over the entire period of our analysis, instead of from all over the tropics as assumed in our input function), this would lead only to a shift in mean-age values, but not to a long-term trend. To test the influence of such a shift in the tropospheric input region, we have estimated the effect of averaging the tropospheric data between 0 and 20° N, respectively 20° S instead of between 20° N and 20° S. We have further tested the influence of a possible seasonal cycle in the input of air into the stratosphere[29], with a maximum flux during Northern Hemisphere winter, by applying an artificial 15% seasonal cycle to the input data. Uncertainties due to the representativeness of the second-order polynomial have also been assessed. We further estimated uncertainties in the early SF_6 tropospheric reference data, which are derived from emissions, by arbitrarily varying these emissions by 10% (1 sigma) in a Monte Carlo simulation. Finally, the uncertainty due to the parameterization of the width of the age spectrum has been calculated for each flight by varying this parameterization between $\Delta^2/\Gamma = 0.5$ and 1.25.

As balloon observations provide only a snapshot of the atmosphere, we investigated the expected variability in the stratosphere, on the basis of trace gases for which a larger data set is available. Typical stratospheric profiles and variabilities of CH_4 have been taken from the UARS reference atmosphere and expected variabilities in mean age have been derived from this for the pressure range between 30 and 5 hPa over the entire year and for the same latitude band as used in the observations. On the basis of a correlation function between mean age and CH_4 derived from the observations between 1995 and 2005, this variability has been transformed into an expected variability in mean age. This variability is expected to be 0.31 years.

All errors and uncertainties were added up to produce a maximum total uncertainty estimate (1 sigma), which was used as an error in the trend calculation. In addition, we have repeated the calculation by systematically excluding each of the data sets used for the trend analysis. Under these circumstances, the smallest trend was +0.2 years per decade (when removing the LACE data), and the largest trend was +0.35 years per decade (when removing the NCAR flights dating back to 1975).

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Additional information

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