scale of cooling. Given comparatively fast cooling, one may even expect that most of the elements condense as single oxides and that they later react with each other to form more complex oxides and silicates. Some of the oxide grains may survive.

Another possibility for the formation of oxide inclusions is grain boundary exsolution within constituents of CAIs if local enrichments of Mg, Ca, Ti, or Al occurred during the crystallization of the host minerals. On the basis of textural observations, such a process seems unlikely. (i) The oxides do not exhibit typical exsolution features. In contrast, some inclusions exhibit euhedral morphologies, which would be unusual for exsolution. (ii) Within an area of 20 nm by 50 nm, three different oxides (TiO_2, CaO, and Al_2O_3) were encountered (Fig. 1C). In the case of diffusion-controlled exsolution, the thermodynamically stable phases $CaTiO_3$ (perovskite) and $CaAl_4O_7$ (grossite) would have formed instead of pure oxides. Because exsolution requires the nucleation of a phase so that the resulting two-phase assemblage is more stable than the parent solid solution, it is unlikely that this process leads to the formation of metastable oxide phases. (iii) The exsolution of MgO and Al_2O_3 from spinel; CaO from spinel, perovskite, and melilite; and TiO_2 from spinels and perovskites would be a very unusual phenomenon with no terrestrial equivalent. The range of oxide phases, and in particular the presence of CaO within three different phases, makes it improbable that the oxides were formed by exsolution.

Excluding exsolution as the formation process, we suggest that these inclusions are primary condensates that were either enclosed within or between the host minerals, mostly spinels, and were therefore protected from being consumed by reactions with the remaining solar gas. However, because such phases are not formed by equilibrium condensation, it will be difficult to decipher their formation in detail.

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13. Samples were ion thinned with a GATAN 600 DIF ion mill at 4.5 kV and 12°.
14. Fassaite in Acfer 082 contains 2.5 to 3.5 weight % TiO_2; in Acfer 086, 2.5 to 4.0 weight %; in Allende, up to 18 weight % (*8*); and in Acfer 094, 2.0 to 4.5 weight %.
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Decline in the Tropospheric Abundance of Halogen from Halocarbons: Implications for Stratospheric Ozone Depletion

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Analyses of air sampled from remote locations across the globe reveal that tropospheric chlorine attributable to anthropogenic halocarbons peaked near the beginning of 1994 and was decreasing at a rate of 25 ± 5 parts per trillion per year by mid-1995. Although bromine from halons was still increasing in mid-1995, the summed abundance of these halogens in the troposphere is decreasing. To assess the effect of this trend on stratospheric ozone, estimates of the future stratospheric abundance of ozone-depleting gases were made for mid-latitude and polar regions on the basis of these tropospheric measurements. These results suggest that the amount of reactive chlorine and bromine will reach a maximum in the stratosphere between 1997 and 1999 and will decline thereafter if limits outlined in the adjusted and amended Montreal Protocol on Substances That Deplete the Ozone Layer are not exceeded in future years.

The Montreal Protocol on Substances That Deplete the Ozone Layer was passed, adjusted, and amended in light of the large body of evidence implicating anthropogenic chlorine- (Cl-) and bromine- (Br-) containing gases in the depletion of stratospheric ozone (O_3) (*1–5*). Human use of halocarbons has led to a steady increase in the amounts of these compounds in the atmosphere for more than 20 years (*6–9*). In the modern atmosphere, relatively few halocarbons emitted from natural processes reach the stratosphere (*2, 3, 5, 10–12*).

Recent reports have highlighted selected anthropogenic halocarbons for which growth rates and abundance have decreased (*6–9*), owing to restrictions outlined in the Montreal Protocol and its subsequent adjustments and amendments (the revised Montreal Protocol). Model studies have predicted that tropospheric Cl will reach a maximum in the mid-1990s at ~3500 to 4000 pmol mol^{-1} [parts per trillion (ppt)] if limits outlined in the Copenhagen Amendments are not exceeded (*3, 13*). These studies further suggest that a minimum for stratospheric O_3 in mid-latitudes will occur 3 to 4 years after the peak in tropospheric Cl and that O_3 abundance will begin to increase as stratospheric Cl concentrations decrease from their maximum. The atmospheric abundance of Br also must be considered, especially because Br is substantially more

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efficient than Cl in destroying stratospheric O_3 (*2, 3, 13*). The abundance of Br relative to Cl in the atmosphere is low (*2, 3, 10*), and restrictions have been implemented in the production and consumption of Br-containing halocarbons—for example, halons and methyl bromide (CH_3Br).

Until now, however, much uncertainty has remained regarding the timing and magnitude of peak halogen loading for the atmosphere. This uncertainty derives from the potential for noncompliance in selected countries, uncertainty in the magnitude of chlorofluorocarbon (CFC) and hydrochlorofluorocarbon (HCFC) use in developing nations, and evidence suggesting significant amounts of illicit trade in CFCs (*14*). Additional emission of halocarbons from these potential sources could enhance stratospheric halogen abundance and O_3 destruction, thus delaying recovery of stratospheric O_3 to preindustrial concentrations.

We show here how current tropospheric abundances of the main anthropogenic halocarbons are likely to affect future halogen concentrations and O_3 destruction in the lower, mid-latitude stratosphere and in the polar stratosphere.

We have determined the abundance and growth rates of CFCs, HCFCs, halons, chlorinated hydrocarbons, and HFC-134a (CF_3CH_2F) in recent time at selected remote locations in both the Northern and the Southern Hemispheres. The halocarbons considered here represent >95% of

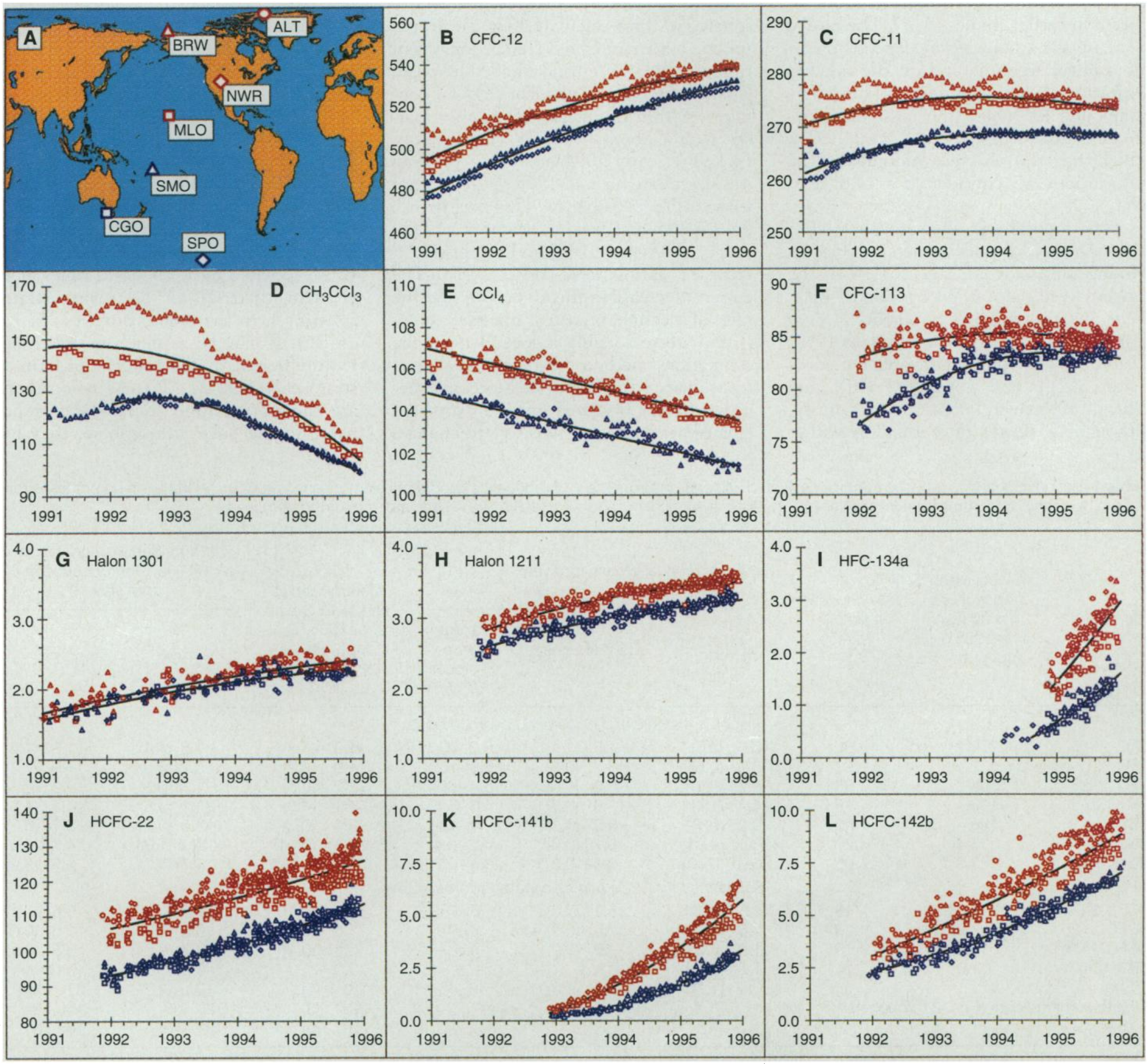

Fig. 1. Atmospheric, dry mole fractions for selected halocarbons at Northern Hemispheric (red symbols) and Southern Hemispheric (blue symbols) sites since 1991. Samples were collected at the sites displayed in (**A**) (*15*). Points represent monthly means for four or five sampling locations as determined by instruments located at those sites (**B** through **E**) or means from flask pairs (**F** through **L**) that had been collected at one of the seven sampling locations and returned to our Boulder laboratory for subsequent analysis (*15*). Solid lines represent fits to monthly hemispheric means, which are not shown for clarity. Provisions within the adjusted and amended Montreal Protocol currently prohibit consumption of molecules displayed in (B) through (H). Units on the ordinate are parts per trillion; sampling dates appear on the abscissa (tick marks correspond to 1 January of indicated year).

total organic Cl and >85% of total organic Br in the atmosphere in 1995 (3, 10, 15). Our analysis shows that, despite uncertainties about the level of emission of these compounds, annual increases in halocarbons for which consumption in developed nations is prohibited by the revised Montreal Protocol have declined in recent years (Fig. 1; Table 1). Furthermore, the tropospheric abundance of certain halocarbons actually decreased in 1995. The decreases we observed for methyl chloroform (CH_3CCl_3) and carbon tetrachloride (CCl_4) extend those reported in earlier studies (8, 9). The rate of decrease for a halocarbon depends on its atmospheric lifetime and on the rapidity with which its emissions are curtailed. The compound for which the most rapid decrease is now being observed is CH_3CCl_3, which has the shortest lifetime of the halocarbons we consider [τ = 4.8 years (9)].

The tropospheric abundance of other halocarbons continues to increase (Fig. 1). Mixing ratios for CFC-12 (CCl_2F_2), the longest lived halocarbon considered here, were increasing at approximately one-third the rate observed in the late 1980s (Table 1). The atmospheric abundances of halons 1211 and 1301 ($CBrClF_2$ and $CBrF_3$, respectively) continue to increase, although at slower rates than in earlier years (16, 17), despite a ban on production since 1994 (18). Also increasing over this period is the atmospheric abundance of certain HCFCs and hydrofluorocarbons (HFCs) (Fig. 1; Table 1) (19, 20). Although both of these classes of compounds are substitutes for CFCs and chlorinated hydrocarbons, the HCFCs contain Cl and have a small potential to deplete O_3. Accordingly, use of HCFCs is allowed only temporarily (until 2040) under the revised Montreal Protocol.

To discern how this changing mix of trace gases will affect O_3 in the stratosphere, we first calculate how the tropospheric burden of Cl has changed in recent time (3, 13). We find that the mean, global, tropospheric abundance of Cl contained within the major CFCs, chlorinated hydrocarbons, HCFCs, and halons peaked in early 1994 at ~3025 ppt and was decreasing in mid-1995 at 25 ± 5 ppt year^{-1} (Fig. 2; Table 1). This decline is a dramatic change from increases of 110 ppt year^{-1} observed in 1989 and 60 ppt year^{-1} recorded during 1990–1992 (2, 3). The current decrease in the Cl burden and the rate of decline reported for each compound are reasonably independent of the estimation method and the analytical technique (15) (Table 1). Moreover, because trends observed in the Northern Hemisphere are most sensitive to changes in emissions (21), they essentially define future accumulation rates in the Southern Hemisphere and in the global mean. In mid-1995, tropospheric Cl was decreasing in the Northern Hemisphere at 30 ppt year^{-1}, more rapidly than the global mean, and had dropped approximately 50 ppt from the observed maximum. After accounting for Cl contributed by methyl chloride (CH_3Cl), other chlorinated hydrocarbons, and less abundant CFCs (3, 10, 11, 22), we estimate that the mean global Cl loading of the troposphere peaked in 1994 at ~3700 ppt.

Stratospheric O_3 depletion is catalyzed by reactive, inorganic forms of Cl and Br. Accordingly, O_3 depletion is less related to the total burden of halocarbon than it is to the amount of halogen released as halocarbon source gases decompose in the stratosphere. Halogen release rates vary over altitude and latitude, however, as does the efficiency for Br, as compared to Cl, to catalyze the destruction of stratospheric O_3 (the "alpha" factor). Whereas Br is estimated to be about 40 times more efficient than Cl for destroying stratospheric O_3 in the polar vortex (2, 3, 13, 23–26), it may be as much as 100 times more efficient in the lower, mid-latitude stratosphere (3, 24, 26). As halocarbons are transported from the tropical tropopause to the polar stratosphere, they be-

Table 1. Recent global tropospheric trends for selected halocarbons from the National Oceanic and Atmospheric Administration Climate Monitoring and Diagnostics Laboratory air monitoring program. All values in parentheses refer to 1 SD of the estimate; n.d., not determined.

Compound	Global mean tropospheric mixing ratio at 1995.5 (ppt year^{-1})	Atmospheric lifetime (3) (years)	Annual growth (ppt year^{-1})			Contribution to annual Cl growth at 1995.5§ (ppt year^{-1})	Contribution to growth at 1995.5 in troposphere (ppt year^{-1})	
			At 1995.5	Estimate from lag‡	Before Montreal Protocol (2, 3)		EECl¶	ECl
CFCs, halons, and chlorinated hydrocarbons								
CFC-11	272	50	−0.6 (0.6)*	−1.5 (1.4)	9.3–10.1	−1.9 (1.8)	−1.5 (1.4)	−1.9 (1.8)
CFC-12	532	102	5.9 (0.6)*	6.7 (0.2)	16.9–18.2	11.9 (1.2)	5.7 (0.6)	11.9 (1.2)
CFC-113	84	85	−0.7 (0.1)†	−0.2 (0.6)	5.4–6.2	−2.2 (0.3)	−1.3 (0.2)	−2.2 (0.3)
CH_3CCl_3	109	5	−14.7 (1.4)*	−13.0 (0.2)	4.8–5.1	−44.1 (4.3)	−38.1 (3.8)	−44.1 (4.3)
CCl_4	103	42	−0.8 (0.1)*	−0.9 (0.1)	1–1.5	−3.1 (0.3)	−2.6 (0.3)	−3.1 (0.3)
Halon 1211	3.4	20	0.11 (0.01)†	0.12 (0.04)	0.2–0.4	0.11 (0.01)	9.9 (1.0)	4.5 (0.5)
Halon 1301	2.3	65	0.07 (0.07)†	0.02 (0.07)	0.4–0.7	0.0	4.7 (4.6)	2.9 (2.9)
Substitutes for CFCs and chlorinated hydrocarbons								
HCFC-22	117	13	5.6 (0.1)†	5.2 (1.0)	5–6	5.6 (0.1)	1.6 (0.03)	5.6 (0.1)
HCFC-142b	7.2	20	1.6 (0.02)†	1.5 (0.2)	n.d.	1.6 (0.02)	0.5 (0.01)	1.6 (0.02)
HCFC-141b	3.5	9	1.9 (0.03)†	1.6 (0.2)	n.d.	3.8 (0.1)	2.2 (0.04)	3.8 (0.1)
HFC-134a	1.6	14	1.2 (0.1)†	n.d.	n.d.	0.0	0.0	0.00
Sum			−0.4 (1.7)	−0.5 (1.8)	43–48	−28 (5)	−19 (6)	−21 (6)
Fit to summed data (Figs. 2 through 4)#						−22 (2)	−23 (6)	−15 (4)
Average						−25 (5)	−21 (8)	−18 (7)

*Growth at 1995.5 estimated from data obtained with instruments located at remote sampling locations and the GC analysis of air sampled in flasks by both electron capture (GC-ECD) and mass spectrometric detection (GC-MS) (15). †Growth at 1995.5 estimated from flask samples only. Flask air analysis performed either by GC-MS or GC-ECD. Reported mean rate and standard deviation for each compound are from fits to data binned by monthly, bimonthly, odd-month, and even-month periods, and by station. ‡Annual growth rate estimated for the period since January 1994 from the difference between global monthly means 12 months apart (for example, December 1995–December 1994; no fitting involved). Reported values are means and standard deviations for differences calculated for all months in 1995. §Calculated for each compound from growth estimate reported under * or † only. ¶Halogen release rates (23, 24) were multiplied by 0.8 to account for the fractional release of CFC-11 in a single pass through the stratosphere. #Reported mean rate and standard deviation are from fits to summed data binned by monthly, bimonthly, odd-month, and even-month periods, although fits displayed in Figs. 2 through 4 are to monthly means only.

come degraded through photolysis and chemical oxidation reactions. Most halocarbons have undergone extensive photolysis by the time they descend into the springtime polar vortex, so that an upper limit to the stratospheric abundance of O_3-depleting gases can be estimated from the total burden of organic Cl and Br that was originally present (*22*, *27*). In contrast, less of each halocarbon has decomposed in the lower, mid-latitudinal stratosphere; estimates for halogen release rates in this region are based on measurements of Cl- and Br-containing gases in the stratosphere (*2*, *23*, *24*). By weighting tropospheric mixing ratios according to relative decomposition rates for individual compounds within different regions of the stratosphere, we can use tropospheric observations to predict how concentrations of stratospheric inorganic halogen are likely to change in future years (*3*, *24*). In the following analysis, we estimate the future abundances of reactive halogen compounds for the lower, mid-latitudinal stratosphere [effective equivalent chlorine (EECl)] and for the springtime polar stratosphere [equivalent chlorine (ECl)] with halogen release rates and alpha factors appropriate for each region (*28*).

The current mix and abundance of halocarbons within the troposphere ultimately will release fewer halogen atoms to the lower, mid-latitudinal stratosphere than in earlier years (Fig. 3). The mean global tropospheric burden of halogen that will become inorganic halogen in the stratosphere reached a maximum in early 1994 and was declining in mid-1995 at 21 ± 8 ppt EECl year^{-1} (Table 1). We calculate similar rates of decrease for EECl in mid-1995, regardless of the estimation method used (Table 1). In the Northern Hemisphere in mid-1995, the EECl content of the air was changing at −35 ppt year^{-1} and had dropped ~40 ppt below the peak maximum.

The actual rate of change for EECl in mid-1995 may be somewhat lower if the atmospheric abundance of CH_3Br has increased since 1992. We have not explicitly included CH_3Br in our analysis because we were not able to accurately estimate atmospheric growth rates for CH_3Br from air samples that had been stored in flasks (*29*). Increases in the atmospheric abundance of CH_3Br can influence the rate of change and the time for the peak maximum we estimate for EECl. However, limits to production outlined in the Copenhagen Amendments and production figures from the major global producers for 1991 and 1992 suggest that anthropogenic CH_3Br emissions may have stabilized in the early 1990s. We consider it unlikely that an increase in CH_3Br over this period would have been large enough to offset the decrease we report here for EECl (*29*).

For a mean transport time between the troposphere and lower, mid-latitude stratosphere of 3 to 4 years (*3*, *23*, *24*, *30*) and an equivalent contribution of northern and southern tropospheric air to the stratosphere, maximum concentrations of inorganic halogen are expected in the lower, mid-latitudinal stratosphere between 1997 and 1998. Modeling studies suggest that when stratospheric mixing ratios of reactive halogenated compounds begin declining, column O_3 abundances at mid-latitudes will begin to recover (*3*). However, because stratospheric O_3 is influenced by other variables such as aerosol loading and temperature, the exact timing will also depend on how these variables change over this period.

To estimate the stratospheric abundance of O_3-depleting gases in the springtime polar vortex, we calculate ECl on the basis of our tropospheric measurements. The current mix and growth rates of these gases in the troposphere will result in lower ECl in the polar stratosphere in the future. In mid-1995, ECl was decreasing at 18 ± 7 ppt year^{-1} (Table 1; Fig. 4). It is unlikely that an increase in atmospheric CH_3Br in recent time would have been large enough to offset this decrease (*29*). Because transport of air from the lower troposphere to the polar stratosphere below ~25 km occurs in 3 to 5 years (*3*, *13*, *25*, *30*), or over a slightly longer period than transport of air to the lower, mid-latitude stratosphere, we expect that ECl will reach a maximum in the polar stratosphere between 1997 and 1999 and will decline thereafter as long as current growth rates for halons and CFC-12 and the abundances of other CFCs and halocarbons continue to decline.

Although the abundance of reactive halogen in the polar stratosphere above Antarctica will decline when air currently within the troposphere reaches this region, the total column O_3 concentrations in springtime will not increase there immediately. Ozone has been nearly completely destroyed in the lower stratosphere above the Antarctic continent in springtime for the past 8 years (*2*, *3*). The total column O_3 abundance within this region will begin

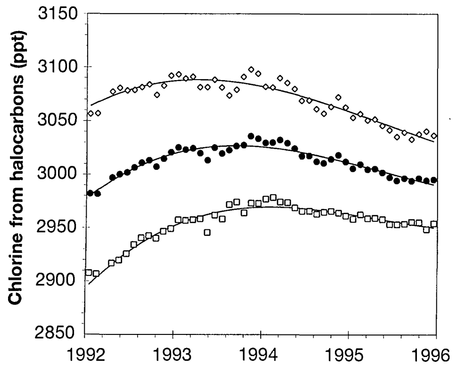

Fig. 2. Tropospheric Cl abundance estimated on a monthly basis for the Northern Hemisphere (open diamonds), the Southern Hemisphere (open squares), and the global mean (filled circles) from the halocarbons shown in Fig. 1. Each point is determined from the sum of monthly hemispheric means for individual halocarbons after accounting for the number of Cl atoms per halocarbon. Solid lines represent fits to monthly means.

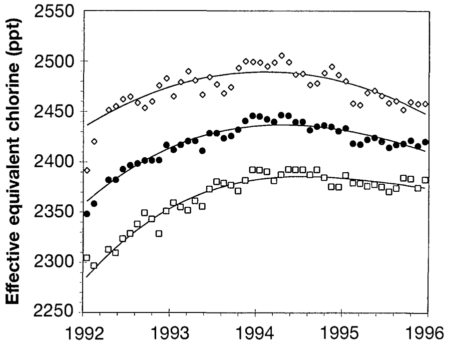

Fig. 3. Amount of Cl and Br (as effective equivalent chlorine or EECl) in the troposphere that is predicted to be released as inorganic halogen in the lower, mid-latitude stratosphere at some future time from the halocarbons shown in Fig. 1. Symbols and lines are as described in Fig. 2. Time on the *x* axis corresponds to when samples were acquired at Earth's surface. These times are expected to be relevant for the lower, mid-latitudinal stratosphere only after adding ~3 to 4 years (see text).

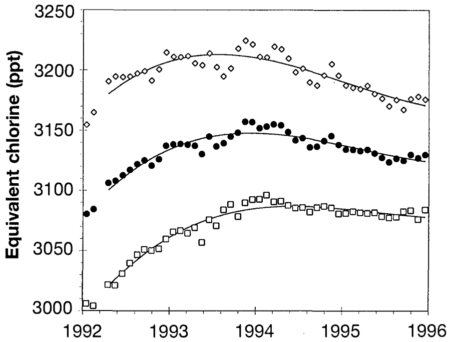

Fig. 4. Amount of Cl and Br (as equivalent chlorine or ECl) in the troposphere that is predicted to be released as inorganic halogen in the springtime, polar stratospheric vortex at some future time from the halocarbons shown in Fig. 1. Symbols and lines are as described in Figs. 2 and 3. Time on the *x* axis corresponds to when samples were acquired at Earth's surface. These times are expected to be relevant for the polar stratospheric vortex only after adding ~3 to 5 years (see text).

to recover only when the concentrations of reactive halogenated compounds drop below those present in the late 1980s (3, 13).

Despite concern that halocarbon production might exceed limits outlined in the revised Montreal Protocol, the current tropospheric mix of halocarbons will eventually release fewer halogen atoms to the stratosphere than in previous years. If current trends continue, we expect stratospheric O_3 mixing ratios to reach a minimum in the near future and in the lower, mid-latitude stratosphere to begin recovering by the turn of the century. However, without widespread adherence to restrictions outlined in the revised Montreal Protocol, additional emissions of Cl- and Br-containing compounds could potentially slow or reverse the trends reported here and delay recovery of stratospheric O_3.

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15. Annual growth for CFC-11 (CCl_3F), CFC-12 (CCl_2F_2), CH_3CCl_3, and CCl_4 was estimated from three different techniques: (i) hourly analysis of air by gas chromatography (GC) with electron capture detection (ECD) at four or five remote sampling locations (*6*): the National Oceanic and Atmospheric Administration Climate Monitoring and Diagnostics Laboratory (NOAA-CMDL) Observatories at Point Barrow, AK (BRW; 71.3°N, 156.6°W); Mauna Loa, HI (MLO; 19.5°N, 155.6°W); Cape Matatula, American Samoa (SMO; 14.3°S, 170.6°W); and the South Pole (SPO; 90°S); and a cooperative site at Niwot Ridge, CO (NWR: 40.1°N, 105.5°W); (ii) collection of flask air samples that were subsequently analyzed by GC with ECD (*6, 16*); and (iii) collection of flasks that were subsequently analyzed by GC with detection by mass spectrometry (MS) (*19*). Flasks were collected in pairs from one to four times per month at the sites listed above and at two additional locations: Alert, Northwest Territories, Canada (ALT; 82.5°N, 62.3°W) and Cape Grim Baseline Air Pollution Station, Tasmania, Australia (CGO: 40.7°S, 144.8°E). Dry-air mole fractions reported for HCFC-22 ($CHClF_2$), HCFC-142b (CH_3CClF_2), HCFC-141b (CH_3CCl_2F), HFC-134a (CF_3CH_2F), and halon 1211 ($CBrClF_2$) were determined with GC-MS detection from air collected in flask pairs (*19*). Those reported for halon 1301 ($CBrF_3$) were determined from these flask samples with a GC-ECD instrument (*16*). Chromatographic peak areas or heights were determined for each halocarbon in air samples and were compared to those obtained in a reference sample of real air. We estimated dry-air mole fractions by comparing reference air to primary standards that had been prepared with gravimetric techniques at mixing ratios similar to ambient air. All data were weighted by the cosine of the sampling latitude before hemispheric means or hemispheric growth rates were estimated. Owing to the redundancy provided by multiple instruments and multiple gas calibration standards, we estimate that the influence of any potential drift within our standards or reference air samples affects the rate of change reported for Cl in mid-1995 by less than 5 ppt $year^{-1}$. Less abundant CFCs not measured here accounted for growth of only ~1 ppt $year^{-1}$ (CFC-114) (*3*) and ~0.45 ppt $year^{-1}$ (CFC-115) (*25*) between 1990 and 1992. This growth probably has diminished since that time as these compounds also are restricted by the Montreal Protocol and its adjustments and amendments.
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27. Some compounds such as HCFC-22, and HCFC-142b undergo slower decomposition in the stratosphere than do most CFCs and are only partially decomposed, even in the most photochemically aged air reaching the lower polar stratosphere (*22*).
28. We calculate, on the basis of our tropospheric halocarbon measurements, what has been defined elsewhere as the effective equivalent stratospheric Cl (EESC) content of air (*3, 24*). However, in our use of this term we consider time lags associated with transport separately, so we label it as effective equivalent chlorine only (EECl). Values for halogen release rates in the lower, mid-latitudinal stratosphere relative to CFC-11 are taken from those tabulated in (*24*). These release rates are estimated from observed or computed relations between individual compounds and CFC-11. Furthermore, we assume that these long-lived gases are well mixed vertically within the troposphere, and that the amount of inorganic halogen released and available to interact with O_3 in the stratosphere is well estimated by the amount of halocarbon that has been destroyed in the stratosphere. All computations involving the lower, mid-latitude stratosphere were made with alpha = 100 (*3, 24, 26*). In the calculation of ECl in the springtime polar vortex, we estimated alpha at 40 (*2, 3, 13, 23–26*).
29. We are not able to determine accurately growth rates for CH_3Br from samples collected and analyzed here, owing to problems associated with the storage of air in flasks [S. A. Montzka *et al.*, *Eos* **76** (no. 17) (spring suppl.), 160 (1995); J. M. Lobert *et al.*, *NOAA Tech. Mem. ERL CMDL-10* (1996)]. However, M. A. K. Khalil *et al.* [*J. Geophys. Res.* **98**, 2887 (1993)] estimated growth for tropospheric CH_3Br at 0.3 ± 0.1 ppt $year^{-1}$ (3% per year) from flask air samples collected in the 4 years before 1992. During this period, anthropogenic production was increasing at 5 to 6% per year. It is not likely that annual increases would have continued at this rate after 1992, as the Copenhagen Amendments to the Montreal Protocol limit production and consumption for CH_3Br beginning in 1995 to the amount produced and consumed in 1991; production was frozen a year earlier (1994) in the United States. Sales of CH_3Br from the major producing companies throughout the globe in 1992 were only 0.5% higher than in 1991 [*Methyl Bromide Annual Production and Sales for the Years 1984–1992*, (Methyl Bromide Global Coalition, Washington, DC, June 1994)]. If production and emission remained constant from 1991 through 1995, we anticipate a mean increase of <0.02 ppt $year^{-1}$ through this period, which is insignificant in our analysis. To estimate an upper limit for the influence of growth in CH_3Br abundance, we consider the increasing emissions after 1992 of 10^3 tons $year^{-1}$ (either from a continued increase in emission or from increases in biomass burning of ~5% per year (M. O. Andreae *et al.*, *J. Geophys. Res.*, in press). We estimate that increasing emissions of this magnitude would result in an annual rate of change in mid-1995 that is ~5 ppt $year^{-1}$ (ECl) or ~10 ppt $year^{-1}$ (EECl) (or 30 to 50%) less negative than what we report, assuming no increase in CH_3Br abundance; the maximum EECl or ECl would have occurred <0.5 year later. As an estimate for global tropospheric burden we use 10 ppt [J. M. Lobert *et al.*, *Science* **267**, 1002 (1995)].
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31. We are indebted to all personnel involved in collecting flask samples at the NOAA/CMDL Observatories and at cooperative sampling sites: P. J. Fraser and staff at the Cape Grim Baseline Air Pollution Station; M. Losleben (University of Colorado); and N. Trivett and staff at Alert Canada (Atmospheric Environment Service). We appreciate useful discussions with J. S. Daniel and the technical assistance of N. J. Condon. This work was supported in part by the Atmospheric Chemistry and Radiatively Important Trace Species components of NOAA's Climate and Global Change Program.

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