UC Irvine

UC Irvine Previously Published Works

Title

Continental sources of halocarbons and nitrous oxide

Permalink

<https://escholarship.org/uc/item/3jr4m5qv>

Journal

Nature, 317(6034)

ISSN

0028-0836

Author

Prather, Mj

Publication Date

1985-12-01

DOI

10.1038/317221a0

Copyright Information

This work is made available under the terms of a Creative Commons Attribution License, available at <https://creativecommons.org/licenses/by/4.0/>

Peer reviewed

15. Sanders, D. B., Solomon, P. M. & Scoville, N. Z. *Astrophys. J.* **276**, 182–203 (1984).
16. Lebrun, F. *et al. Astrophys. J.* **274**, 231–236 (1983).
17. Bhat, C. L., Issa, M. R., Houston, B. P., Mayer, C. J. & Wolfendale, A. W. *Nature* **314**, 511–515 (1985).
18. Bahcall, J. N. & Soneira, R. M. *Astrophys. J. Suppl.* **44**, 73–110 (1980).
19. Hertz, P. & Grindlay, J. E. *Astrophys. J.* **278**, 137–149 (1984).
20. Tammann, G. A. in *Supernovae: A Survey of Current Research* (eds Rees, M. J. & Stoneham, L. R. J.) 371 (Reidel, Dordrecht, 1982).
21. Lyne, A. G., Manchester, R. N. & Taylor, J. H. *Mon. Not. R. astr. Soc.* **213**, 613–639 (1985).
22. Webber, W. R., Simpson, G. A. & Cane, H. V. *Astrophys. J.* **236**, 448–459 (1980).
23. Baldwin, J. E. in *The Structure and Content of the Galaxy and Galactic Gamma Rays* (NASA CP-002, 1976).
24. Strong, A. W. *Mon. Not. R. astr. Soc.* **181**, 311–322 (1977).
25. Kniffen, D. A. & Fichtel, C. E. *Astrophys. J.* **250**, 389–397 (1981).
26. Koyama, K. in *X-ray Astronomy '84* (eds Oda, M. & Giacconi, R.) 325–328 (1984).
27. Shapiro, P. R. & Field, G. D. *Astrophys. J.* **205**, 762–765 (1976).

Continental sources of halocarbons and nitrous oxide

Michael J. Prather*

Center for Earth and Planetary Physics, Harvard University, Cambridge, Massachusetts 02138, USA

Estimates of continental sources of CFC-11, CFC-12, CCl₄, CH₃CCl₃ and N₂O are derived from the atmospheric lifetime experiment in Adrigole, Ireland, and anthropogenic emissions of CCl₄ and N₂O from Europe have been identified. Relative source strengths are consistent with global budgets for the halocarbons and N₂O. Different industrial release patterns for halocarbons are observed for Europe, the western United States and Australia.

RECORDS of the composition of the atmosphere over the past two decades^{1–9} show a global increase in the concentrations of CO₂, CH₄, N₂O, CO, chlorofluorocarbons-11 and -12 (CFC-11 and CFC-12) as well as many other halocarbons. Observations have often focused on clean-air, baseline conditions where a reliable trend in the global burden of these gases may be determined. Here, I analyse data for trace gases from a once polluted site (Adrigole, Ireland) to identify and quantify sources of N₂O and halocarbons from the European continent. Globally, these gases represent the major stratospheric supply of odd nitrogen and inorganic chlorine, which, in turn, control the abundance of ozone. Increasing levels of chlorine and odd nitrogen may lead to large reductions in stratospheric ozone^{10,11}.

Sources of atmospheric trace compounds such as N₂O and halocarbons are poorly quantified. Emission inventories for halocarbons are derived through surveys and estimates of global manufacture and distribution which contain major uncertainties^{9,12}. The budget for N₂O (ref. 7) is based on calculated losses from the stratosphere, on global scaling of few measurements of fluxes from soils and oceans, on the observed rate of increase in atmospheric concentration and on the detection of N₂O as a by-product of combustion^{13,14}. I derive here an accurate calibration of the relative emission of N₂O and halocarbons from Europe. This information is contained in the measurements of CFC-11 (CFCl₃), CFC-12 (CF₂Cl₂), CCl₄, CH₃CCl₃ and N₂O from the atmospheric lifetime experiment (ALE) station at Adrigole, Ireland⁹.

The ALE data have been presented in the form of monthly mean concentrations from which the pollution events have been removed⁹. The initial study was extensive and used these clean-air, baseline values to calculate highly accurate hemisphere and global trends to compare with estimates for industrial emissions, and thence to derive globally averaged atmospheric lifetimes. I examine here the variations of trace gas concentrations occurring on short timescales, from 1 to 10 days, which result from the passage of polluted air over the observing site. The complete set of individual observations from the first 3 yr at Adrigole has recently become available and is shown in Fig. 1. The ALE programme has noted that ~30% of these measurements at Adrigole seem to be polluted, containing significantly higher concentrations of specific halocarbons associated with anthropogenic sources in Europe. The ALE analysis has selectively removed these events; here, I focus on them, using the elevated concentrations during pollution events to derive relative source strengths of these gases from western Europe. Similar,

but less detailed, analyses have been reported for urban sources of methane¹⁵ and for CFCs in Australia (P. J. Fraser, personal communication). The site at Adrigole is particularly advantageous for sampling European land sources with respect to the relatively uniform oceanic background. ALE data from Cape Meares, Oregon and Cape Grim, Tasmania do not define continental emissions as clearly as the Adrigole data, but do indicate differing relative sources of these gases from western Europe, North America and Australia.

ALE measurements and analysis

The first 3 yr of measurements from the Adrigole station, spanning 1 July 1978 to 30 June 1981, are presented in Fig. 1. The gas chromatograph used two different columns, silicone and porosil, and values for CFC-11, designated S and P, were reported for both⁹. The data show numerous pollution episodes which seem to correlate across all gases except N₂O. Irregular intervals in the record complicate the time-series analysis.

In the current analysis, we have processed the ALE data for each of the 'six' gases individually as follows:

- (1) Original measurements, taken several times a day, are recorded to the nearest minute of observations, and have been temporally degraded into intervals of 8 h; where more than one observation falls within the 8-h interval, they are averaged.
- (2) A linear, least-squares regression was fitted to all of the 8-h data; all points were weighted equally and no attempt was made to fill gaps in the record.
- (3) These 'detrended' data were sorted, resulting in the probability distribution functions shown in Fig. 2.
- (4) A limit on extreme values was selected from the 0.5% and 99.5% probability levels; and those data in the upper/lower 0.005 fractile were rounded down/up to the chosen extrema.
- (5) The truncated data set was again fitted with a linear trend which was again removed.
- (6) Auto-correlations and cross-covariances of the residual variances were computed with a resolution of 8 h; again, no attempt was made to fill the numerous gaps in the data. Covariances (and auto-correlations) have been defined as

$$C_{12}(\Delta) = \frac{1}{N} \sum f_1(t) \times f_2(t + \Delta)$$

where there are N such pairs of residual trace gas mixing ratios, f_1 and f_2 , available at a time lag Δ .

(7) The coherence of a pair of gases (1, 2) is defined in the form of a normalized covariance,

$$h_{12}(\Delta) = c_{12}(\Delta) / [c_{11}(\Delta) \times c_{22}(\Delta)]^{1/2}$$

where the relative covariances (c_{12}) represent the increase in

* Present address: NASA/Goddard Institute for Space Studies, 2880 Broadway, New York, New York 10025, USA.

Fig. 1 ALE measurements from Adrigole, Ireland. All primary observations from the P-column (CFC-11, CFC-12, N_2O) and the S-column (CFC-11, CH_3CCl_3 , CCl_4) which fall within the ranges of the graphs have been plotted. Dashed lines indicate the 99-percentile level.

variance of short-term pollution events with respect to a baseline defined by the 20–30-day lag.

$$c_{12}(\Delta) = C_{12}(\Delta) - \langle C_{12} \rangle_{20-30\text{-day}}$$

This definition of coherence is meaningful only when C is significantly greater than the baseline, and thus values in Table 2 are reported only for lags out to 8 days where the pollution events are clearly distinguishable above the baseline concentrations of clean air.

Steps 4–5 were adopted because variances in the halocarbon data tend to be dominated by relatively few observations with very high values. This approach effectively filters the extreme, short-lived, infrequent pollution events into manageable variations in trace gas concentrations, without losing the basic information that the air was highly polluted. Repeating this analysis with limits placed at the 1.5 and 98.5% probability levels shows negligible sensitivity to these limits.

Trends in concentration for the trace gases as calculated in steps (2) and (5) are shown together with results from the original ALE analysis in Table 1. Except for CFC-11S these trends agree well and indicate that inclusion of the pollution events (removed by the ALE investigation in the original analysis) do not seem to distort the baseline trend observed at Adrigole. The calculated trend in CFC-11S is influenced by a few extremely high concentrations early in the record: truncating the distribution at $\pm 1.5\%$ probability raises the calculated trend to $+4.6 \pm 0.3\% \text{ yr}^{-1}$, closer to the other trends derived for CFC-11.

The cumulative probability distributions shown in Fig. 2 reveal two distinct categories of data. The linear region below the 60% level is indicative of a normal gaussian distribution that we assume represents the measurement of background air as influenced by instrumental variations plus short-term and seasonal variations not removed by the linear trend. Second, the steep rise in concentrations at the upper quartile of the probability distribution (for all gases but N_2O) points to sources of air with highly enriched concentrations, agreeing with the ALE observation that Adrigole experiences polluted European air $\sim 30\%$ of the time. These distributions may be used to derive relative source strengths for these gases as described below.

The temporal correlation of variations in the five ALE trace gases provide valuable information on their concurrency as well as on the duration of pollution events. The former is addressed here as we attempt to extract relative emissions integrated over a continent; the latter provides important tests for atmospheric models of tracer transport. Our focus is restricted to ± 30 -day lags because of interest in synoptic-scale variability. The original data exhibit discontinuities on a monthly or seasonal basis which make the interpretation of longer correlations more difficult (see

Fig. 2 Probability-distribution functions for the ALE data from Adrigole, Ireland. The primary data shown in Fig. 1 have been sorted into intervals of 8 h, and a least-squares, linear trend has been removed.

Table 1 ALE data from Adrigole

Gas	Column	Factor	ALE analysis		Present analysis	
			Mean	Trend (% yr ⁻¹)	Mean truncated/full	Trend (% yr ⁻¹)
CFC-11	P	0.96	178.8	+5.1 ± 0.2	184.6/184.9	+4.8 ± 0.3
CFC-12	P	0.95	301.3	+6.3 ± 0.2	307.1/307.4	+6.0 ± 0.2
N ₂ O	P				306.0/306.0	-0.2 ± 0.5
CFC-11	S	0.96	177.9	+5.0 ± 0.2	184.2/184.5	+4.4 ± 0.3
CH ₃ CCl ₃	S	0.80	152.3	+7.7 ± 0.6	161.6/161.9	+7.9 ± 0.7
CCl ₄	S	0.80	124.6	+2.7 ± 0.6	126.2/126.4	+2.8 ± 0.4

Mean values refer to January 1980 and have units of parts per 10¹² by volume, except for N₂O with units of parts per 10⁹ by volume. Suggested calibration factors have not been applied (see ref. 9). ALE analysis does not include the 'pollution events', ~30% of observations. Reported trends are for the truncated data set; those for the full set differ by less than the quoted error (95% confidence range for the linear fit).

Table 2 Coherences with respect to CFC-11S

±lag (h)	CFC-11P	CFC-12	N ₂ O	CH ₃ CCl ₃	CCl ₄
0	0.92	0.89	0.19	0.86	0.56
8-24	1.00	0.97	0.28	0.94	0.71
32-104	1.01	0.98	0.35	0.99	0.90
112-176	1.00	0.97	0.32	0.95	1.08

Values are based on the initial (linear-trend) analysis described in text.

Fig. 1). Autocorrelations for the six ALE gases are shown in Fig. 3; and selected cross-covariances, in Figs 4 and 5.

Trace gas correlations

All gases with the exception of N₂O (that is, the halocarbons) exhibit similar patterns: the full width of a pollution event is 2 days at half height; enhanced concentrations persist for long periods (autocorrelations of 20% peak variance extend out to ±5 days); a significant repetition of these events occurs on a scale of 5-8 days with additional structure out to 30 days. Air containing elevated concentrations of halocarbons (such as, variances of 200 (parts per 10¹² by volume, p.p.t.)² is clearly identifiable against the mean background (such as, variances of 10 (p.p.t.)² derived from the probability distribution in Fig. 2). Both N₂O and CCl₄ data are significantly, positively correlated at ±30 days, a consequence of seasonal/annual variability not removed by a linear trend, or possibly a result of their relatively shorter data records (see Fig. 1). The different halocarbons are indeed highly correlated with each other as may be seen in Fig. 4, where the cross-covariances show the same patterns evidenced in the autocorrelations.

For N₂O the uncorrelated noise in the autocorrelation at a time lag of zero, presumably the instrumental variance, is much greater than the correlated variance for lags greater than zero. Its magnitude, ~5 (parts per 10⁹ by volume, p.p.b.)², is consistent with that derived from the slope of the probability distribution, ~7 (p.p.b.)². Furthermore, the N₂O observations show a broad autocorrelation of the order of 1.5 p.p.b., extending over a range of ±15 days. Examination of the N₂O observations in Fig. 1 points to systematic shifts in concentration every calendar month that might lead to such a correlation extending over 30 days. Because of these problems with N₂O, I have re-examined the original data with a different analysis, replacing steps (2)-(6) with the following:

- (2') Reference values for the 8-h data are selected on a month-by-month basis, setting the baseline at the 50th percentile (that is, median) of observations for the month.
- (3') The data are sorted as before.
- (4') The ±0.5% extreme values are truncated as before.
- (5') In a further effort to reduce noise, the apparently aberrant

Fig. 3 Autocorrelations of the six ALE gases from Adrigole. The filled square denotes the zero-lag autocorrelation (white noise) of the time series. Scale factors for the variances are given in each panel. The upper six panels refer to time series from which the linear trend has been removed; the lower two panels (*) show results from the monthly median analysis.

N₂O observations from March through June of 1980 have been dropped (see Fig. 1).

(6') Correlations and covariances are computed as before; selected correlations from this monthly median analysis are included in Figs 3 and 5, denoted by asterisks.

This second analysis does not significantly alter the halocarbon self-correlations, but it greatly reduces the noise in the N₂O time series. As expected, when the data are renormalized by monthly medians, autocorrelations at ±30 days are nearly zero. The covariance of N₂O with the other halocarbons is now more readily visible, and the uncorrelated noise in the data has been reduced to 2.5 (p.p.b.)². The N₂O signal which correlates with pollution events is small, <0.5 p.p.b., and care is needed to ensure that it is not caused by interference in the gas chromatograph with a pollution-associated trace gas such as CO₂. The coherence of N₂O with respect to CFC-11S now exceeds 0.4 at 5-8-day lags.

No pair of gases is perfectly coherent at zero lag, not even CFC-11S and CFC-11P. This may be understood in that a time lag of zero does not mean that both S and P columns sampled the same air mass, but rather that both measurements were made within 8 h of each other. Beginning at a lag of ±8 h and extending out to ±8 days the major halocarbons (CFC-11S, CFC-11P, CFC-12 and CH₃CCl₃) become highly coherent (0.98 ± 0.03), indicating that pollution events of duration >8 h have evolved from quantitatively comparable sources of these gases. Carbon tetrachloride, on the other hand, is not fully coherent with the other halocarbons (for example, 0.71 at 8-24 h) until we reach lags of 5-8 days. The asymmetry of the CCl₄ cross-covariances would imply that elevated levels of CCl₄ are likely to precede a general halocarbon pollution event by about 10 days. A preliminary interpretation of this pattern would be that the major CCl₄ sources observed at Adrigole are indeed European but of spatially distinct regions (for example, eastern Europe) as compared with those of major CFC sources (for example, western Europe).

Fig. 4 Cross-covariances of the halocarbons at Adrigole. The covariances have been defined in the sense given by the formula in the text. Each panel denotes the cross of $f_1 \times f_2$ with the solid line referring to positive lags ($+\Delta$) and the dashed line to negative lags ($-\Delta$). Scale factors for the variances are included in each panel.

Sources

European source strengths for CFC-12, CH_3CCl_3 , CCl_4 and N_2O with respect to that for CFC-11 are derived by two methods and reported in Table 3. We may determine relative emissions from the probability distributions if the highest concentrations observed for each gas are assumed to be perfectly correlated, such that the difference in concentrations between, for example, the 90 and 98% levels is proportional to the source. Molar ratios of these inferred sources are calculated from the probability distributions in Fig. 2 with the background (variance) removed. Relative sources of trace gas 2 with respect to reference gas 1 are derived from the time-series by averaging the ratio of the relative covariance ($c_{12}(\Delta)$) to the autocorrelations of the refer-

Fig. 5 Cross-covariances of N_2O with halocarbons at Adrigole. The covariances are defined as in Fig. 4. The upper panels refer to the analysis with linear trends removed; the lower four panels (*) are based on the monthly median analysis with some of the N_2O data excluded (see text).

ence gas ($c_{11}(\Delta)$). Averages are based on time lags >8 h and <8 days; contemporaneous measurements (zero lag) are not used because their coherence is low and the instrumental noise may be greater than the atmospheric signal (for example, N_2O at Adrigole, CFCs at Cape Meares).

Nitrous oxide sources have been estimated with respect to all the halocarbons and span more than a factor of 2 as indicated. Because the halocarbons are so highly coherent, estimates based on the probability distributions alone agree with the more accurate time-series analyses. Table 3 also includes results of similar analyses for the ALE data from Cape Meares, Oregon and Cape Grim, Tasmania. These latter two stations observe pollution events of smaller magnitude less frequently than Adrigole, requiring a time-series analysis to characterize accurately the atmospheric events above instrumental noise. Unfortunately, no estimates of N_2O sources may be inferred from the Oregon or Tasmania data.

These derived emissions are independent of the local meteorology and synoptic processes as long as the variability at Adrigole results from addition of these inert tracers to a relatively well mixed, uniform background atmosphere. Meteorological studies of halocarbon pollutions at Adrigole^{9,10} clearly point to air that has travelled over the European continent and the British Isles. An absolute calibration of the halocarbon emissions would require not only knowledge of meteorological conditions, but also a proven model for boundary layer mixing and atmospheric transport. Nevertheless, case studies of the CCl_4 episodes may enable positive identification of this source with eastern Europe.

Conclusions

Release patterns for the CFCs are similar for Europe and the western United States, although Australian releases of CFC-12 with respect to CFC-11 are noticeably smaller. Most importantly, global budgets for CFCs^{9,12}, based on observed atmospheric trends and on industrial inventories, indicate a substantially larger ratio of CFC-12 emission to CFC-11 emission (~ 1.8) than is observed for all three regions studied here (~ 1.1). The Soviet Union and developing countries have been proposed as playing a relatively more important role in world use of CFC-12. The Chemical Manufacturers Association (CMA) estimates¹² of the CFC-12: CFC-11 ratio for reporting (western) companies (~ 1.5)

Table 3 Relative molar source strengths

	CFC-11	CFC-12	CH_3CCl_3	CCl_4	N_2O
Probability distribution*					
ALE (Ireland)	1	1.0 ± 0.1	1.9 ± 0.2	0.23 ± 0.07	-0 ± 100
Time series†					
ALE (Ireland)	1	1.1	1.8	0.28	30–70
ALE (Oregon)	1	1.2 ± 0.2	2.5 ± 0.3	0.15 ± 0.05	ND
ALE (Tasmania)	1	0.9 ± 0.2	0.9 ± 0.2	0.15 ± 0.04	ND
Industrial emissions‡					
Global (flux in 10^9 yr^{-1})	1 (265)	1.76 (410)	1.94 (500)	0.34 (100)	52 (2,800 of N)

* Dimensionless ratios are based on the probability distribution between the 90th and 98th percentile ranking; error estimates are subjective, including uncertainties in determination of the high-end probability distributions.

† Error estimates are $\pm 10\%$ for all but N_2O (see text). Absolute calibration factors for the ALE data have not been applied, and would reduce the relative sources for CCl_4 and CH_3CCl_3 by $\sim 15\%$. N_2O sources for Oregon and Tasmania are not detectable (ND).

‡ Estimates for industrial emissions are generally available only for the entire Northern Hemisphere (see refs 9, 12). N_2O flux estimate corresponds to $0.2\% \text{ yr}^{-1}$.

is lower than the global average but still greater than observed for the United States and Europe.

Emissions of CH_3CCl_3 (with respect to CFC-11) are clearly different from the three continents: the western United States is a factor of 1.3 above the global mean, and Australia a factor of 2 below. A balance of sources from the United States and western Europe could maintain the CH_3CCl_3 global budget.

Carbon tetrachloride shows a large European source (with respect to CFC-11) which is still less than estimates for the mean global source. As the United States and Australia seem to contribute lesser relative amounts of CCl_4 , I conclude that additional sources may be required, perhaps also associated with the missing CFC-12. The primary pollution events at Adrigole show a CCl_4 :CFC-11 ratio typical of the other continents (~ 0.15). The larger sources of CCl_4 are associated with high coherence at time lags or more than 4 days, enough time for air to have crossed the European continent.

Nitrous oxide variability at Adrigole shows a component clearly correlated with halocarbon pollution events. This

relationship is consistent with an industrial source of N_2O from combustion of fuel nitrogen^{13,14} which is spatially associated with halocarbon releases. If scaled to global CFC-11 emissions, the magnitude of this source is $\sim 3 \times 10^{12} \text{ g(N) yr}^{-1}$, is equivalent to the observed annual increase in N_2O , and accounts for $\sim 25\%$ of the current global emissions⁷.

Concurrent atmospheric measurements, made at a frequency greater than one per day, provide valuable information on the location and magnitude of sources for many trace gases of both natural and anthropogenic origin. These data permit integration of emissions over large areas and present critical tests for the synoptic-scale transport of trace gases in three-dimensional models. Locations of observing stations near, but not in the middle of suspected sources would allow determination of baseline trends¹⁷ as well as emissions.

This research was supported by NSF grant ATM-81-17009 and NASA grant NAGW-359. I gratefully acknowledge availability of the atmospheric measurements from the ALE team and helpful discussions with S. Wofsy and R. Prinn.

Received 28 March; accepted 10 July 1985.

1. Keeling, C. D. *et al.* *Tellus* **28**, 538-551 (1976).
2. Dianov-Klovov, V. I. & Yurganov, L. N. *Tellus* **33**, 262-273 (1981).
3. Rasmussen, R. A. & Khalil, M. A. K. *Atmos. Environ.* **15**, 883-886 (1981).
4. Ehhalt, D. H., Zander, R. J. & Lamontagne, R. A. *J. geophys. Res.* **88**, 8442-8446 (1983).
5. Craig, H. & Chou, C. C. *Geophys. Res. Lett.* **9**, 477-481 (1982).
6. Khalil, M. A. K. & Rasmussen, R. A. *Science* **224**, 54-56 (1984).
7. Weiss, R. F. *J. geophys. Res.* **86**, 7185-7196 (1981).
8. Fraser, P. J., Hyson, P., Enting, I. G. & Perman, G. I. *Nature* **302**, 692-695 (1983).
9. Prinn, R. G. *et al.* *J. geophys. Res.* **88**, 8353-8441 (1983).

10. *Causes and Effects of Changes in Stratospheric Ozone: Update 1983* (National Research Council, National Academy, Washington DC, 1984).
11. Prather, M. J., McElroy, M. B. & Wofsy, S. C. *Nature* **312**, 227-231 (1984).
12. *Production, Sales and Calculated Release of CFC-11 and CFC-12 Through 1982* (Chemical Manufacturers Association, Washington DC, 1983).
13. Weiss, R. F. & Craig, H. *Geophys. Res. Lett.* **3**, 751-753 (1976).
14. Pierotti, D. & Rasmussen, R. A. *Geophys. Res. Lett.* **3**, 265-267 (1976).
15. Blake, D. R., Woo, V. H., Tyler, S. C. & Rowland, F. S. *Geophys. Res. Lett.* **11**, 1211-1214 (1984).
16. Pack, D. H., Lovelock, J. E., Cotton, G. & Curthoys, C. *Atmos. Environ.* **11**, 329-344 (1977).
17. Graedel, T. E. & McRae, J. E. *Geophys. Res. Lett.* **11**, 977-979 (1980).

Segmentation of mid-ocean ridges

Hans Schouten*, Kim D. Klitgord† & John A. Whitehead*

* Woods Hole Oceanographic Institution, Woods Hole, Massachusetts 02543, USA

† US Geological Survey, Woods Hole, Massachusetts 02543, USA

Studies of mid-ocean ridges in the Pacific and Atlantic oceans show that the volcanism that forms the oceanic crust along the spreading-plate boundaries is concentrated at regular intervals related to spreading rate. This observation and a new calculation for a Rayleigh-Taylor type of gravitational instability of a partially molten mantle region growing under spreading centres yield reasonable estimates of upper mantle viscosities.

SEGMENTATION of mid-ocean ridges seems to be a global phenomenon. Mid-ocean ridges are the spreading boundaries or spreading centres between lithospheric plates where new oceanic crust and lithosphere is created at rates of 10-200 km Myr^{-1} . Although continuous overall, mid-ocean ridges are cut by many transform zones and associated fracture zones that offset the spreading centre by distances up to several hundred kilometres (Fig. 1). These fracture zones divide the ridges into a series of relatively straight segments with lengths of several tens to several hundreds of kilometres. As more high-resolution geophysical surveys of mid-ocean ridges are becoming available in the world's ocean basins, an even more orderly pattern of the sea floor is emerging. This pattern indicates that the boundaries between the spreading plates have a fairly rigorous cellular structure on a 30-80 km length scale.

The spreading centre cells seem to be the primary feature of mid-ocean ridges; they suggest a three-dimensional, rather than two-dimensional, process of magmatism and crustal accretion spaced evenly along the present spreading boundary. Magnetic studies of older oceanic crust suggest relatively stable and even segmentation of plate boundaries in the past on the same length scale^{1,2}. Seismic studies indicate that oceanic crust with normal seismic structure is formed in the spreading centre cells and that between the cells, thinner crust and other structural anomalies

characteristic of fracture zones are formed, regardless of the spatial offset between the spreading centres³⁻⁷.

The transition zones or transform zones between centres on the active mid-ocean ridges manifest themselves in variable and complex ways. Where large offsets exist between the centres, well-defined, predictable transform fault zones provide the continuity and stability of the active plate boundary between the spreading centres on either side of the transform. Where no offsets are apparent between the spreading centre cells, the transition zones between centres are complex, variable and inherently unstable on an approximate length scale of 10 km and timescale of 10^5 - 10^6 yr (refs 8, 9), but they are apparently stable when observed on longer length- and timescales^{1-3,7}.

The apparent stability in space and time on the longer length- and timescales of a regular segmentation of mid-ocean ridges has been attributed by Whitehead, Dick and Schouten¹⁰ to a Rayleigh-Taylor type of gravitational instability of a partially molten mantle region beneath spreading centres. In this model, the wavelength of the instability controls the aggregation and diapiric ascent of mantle melt (magma) at regular intervals along the boundary between two spreading plates.

Here we use the results of high-resolution geophysical surveys of mid-ocean ridges and determine the length scales of segmentation as a function of spreading rate. We use a modification of