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Attention: Dr. Michael Kurylo, Manager

Upper Atmosphere Research Program

Office of Earth Science, Code YS

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entitled

ADVANCED GLOBAL ATMOSPHERIC GASES EXPERIMENT (AGAGE): MIT CONTRIBUTION

for the four year period
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Principal Investigator

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The accomplishments of the Advanced Global Atmospheric Gases Experiment (AGAGE) over the last 4 years have been substantial and documented in a large number of peer-reviewed publications and recorded in data publicly archived for the period June 1978-March 2003 at national and international data centers.

In the following sections we provide, for the 1999-2003 time period, a summary of selected recent research highlights, a complete listing of all central AGAGE publications, a review of AGAGE contributions to international and national assessments, and a list of other publications by the AGAGE team relevant to the project.

(1) RECENT (1999-2003) RESEARCH HIGHLIGHTS

In this section we present abstracts of papers representative of the large number of AGAGE-related papers listed later in Sections (2) and (5). Full references for these abstracts are provided in these two Sections.

A history of chemically and radiatively important gases in air deduced from ALE/GAGE/AGAGE, *J. Geophys. Res.*, 2000

R.G. Prinn, R.F. Weiss, P.J. Fraser, P.G. Simmonds, D.M. Cunnold, F.N. Alyea, S. O'Doherty, P. Salameh, B.R. Miller, J. Huang, R.H.J. Wang, D.E. Hartley, C. Harth, L.P. Steele, G. Sturrock, P.M. Midgely, and A. McCulloch

ABSTRACT. We describe in detail the instrumentation and calibrations used in the ALE, GAGE and AGAGE experiments and present a history of the majority of the anthropogenic ozone-depleting and climate-forcing gases in air based on these experiments. Beginning in 1978, these three successive automated high frequency *in-situ* experiments have documented the long-term behavior of the measured concentrations of these gases over the past twenty years, and show both the evolution of latitudinal gradients and the high frequency variability due to sources and circulation. We provide estimates of the long-term trends in total chlorine contained in long-lived halocarbons involved in ozone depletion. We summarize interpretations of these measurements using inverse methods to determine trace gas lifetimes and emissions. Finally, we provide a combined observational and modeled reconstruction of the evolution of chlorocarbons by latitude in the atmosphere over the past sixty years which can be used as boundary conditions for interpreting trapped air in glaciers and oceanic measurements of chlorocarbon tracers of the deep oceanic circulation. Some specific conclusions are: (a) International compliance with the Montreal Protocol is so far resulting in chlorofluorocarbon and chlorocarbon mole fractions comparable to target levels, (b) Mole fractions of total chlorine contained in long-lived halocarbons (CCl_2F_2, CCl_3F, CH_3CCl_3, CCl_4, $CHClF_2$, CCl_2FCClF_2, CH_3Cl, CH_2Cl_2, $CHCl_3$, CCl_2=CCl_2) in the lower troposphere reached maximum values of about 3.6 ppb in 1993 and are beginning to slowly decrease in the global lower atmosphere, (c) The chlorofluorocarbons have atmospheric lifetimes consistent with destruction in the stratosphere being their principal removal mechanism, (d) Multi-annual variations in chlorofluorocarbon and chlorocarbon emissions deduced from ALE/GAGE/AGAGE data are consistent approximately with variations estimated independently from industrial production and sales data where available (CCl_2F_2 (CFC-12) and CCl_2FCClF_2 (CFC-113) show the greatest discrepancies), (e) The mole fractions of the hydrochlorofluorocarbons and

hydrofluorocarbons, which are replacing the regulated halocarbons, are rising very rapidly in the atmosphere but, with the exception of the much longer manufactured $CHClF_2$ (HCFC-22), they are not yet at levels sufficient to contribute significantly to atmospheric chlorine loading. These replacement species could in the future provide independent estimates of the global weighted–average OH concentration provided their industrial emissions are accurately documented, (f) In the future, analysis of pollution events measured using high frequency *in-situ* measurements of chlorofluorocarbons and their replacements may enable emission estimates at the regional level which, together with industrial end-use data, are of sufficient accuracy to be capable of identifying regional non-compliance with the Montreal Protocol.

Evidence for substantial variations of atmospheric hydroxyl radicals in the past two decades, *Science,* 2001

R. G. Prinn, J. Huang, R. F. Weiss, D. M. Cunnold, P. J. Fraser, P. G. Simmonds, A. McCulloch, C. Harth, P. Salameh, S. O'Doherty, R. H. J. Wang, L. Porter, and B. R. Miller

Abstract. The hydroxyl radical (OH) is the dominant oxidizing chemical in the atmosphere. It destroys most air pollutants and many gases involved in ozone depletion and the greenhouse effect. Global measurements of 1,1,1-trichloroethane (CH_3CCl_3, methyl chloroform) provide an accurate method for determining the global and hemispheric behavior of OH. Measurements show that CH_3CCl_3 levels rose steadily from 1978 to reach a maximum in 1992 and then decreased rapidly to levels in 2000 that were lower than the levels when measurements began in 1978. Analysis of these observations shows that global OH levels were growing between 1978 and 1988, but the growth rate was decreasing at a rate of $0.23 \pm 0.18\%$ year^{-2}, so that OH levels began declining after 1988. Overall, the global average OH trend between 1978 and 2000 was $-0.64 \pm 0.60\%$ year^{-1}. These variations imply important and unexpected gaps in current understanding of the capability of the atmosphere to cleanse itself. [*These conclusions depend on the validity of industry estimates of emissions- see Section 9.3 for further discussion.*]

In situ measurements of atmospheric methane at GAGE/AGAGE sites during 1985 to 2000 and resulting source inferences, *J. Geophys. Res.*, 2002

D.M. Cunnold, L.P. Steele, P.J. Fraser, P.G. Simmonds, R.G. Prinn, R.F. Weiss, L.W. Porter, R.L. Langenfelds, H.J. Wang, L.Emmons, X.X. Tie, and E.J. Dlugokencky

Abstract. Continuous measurements of methane since 1986 at the GAGE/AGAGE surface sites are described. The precisions range from approximately 10 ppb at Mace Head, Ireland during GAGE to better than 2 ppb at Cape Grim, Tasmania during AGAGE (i.e. since 1993). The measurements exhibit good agreement with coincident measurements of air samples from the same locations analyzed by CMDL except for differences of approximately 5 ppb before 1989 (GAGE lower) and about 4 ppb from 1991 to 1995 (GAGE higher). These results are obtained before applying a factor of 1.0119 to the GAGE/AGAGE values to place them on the Tohoku University scale. The measurements combined with a 12 box atmospheric model and an assumed atmospheric lifetime of 9.1 years indicates net annual emissions (emissions - soil sinks) of 545 Tg CH_4 with a variability of only ±20 Tg from 1985 to 1997 but an increase in the emissions in 1998 of 37 ±10 Tg. The effect of OH changes inferred in Prinn *et al.* (2001) is to increase the estimated methane emissions by approximately 20 Tg in the mid-1980s and to reduce them by 20 Tg in 1997 and by more thereafter. Using a 2D, 12 box model with transport constrained by the GAGE/AGAGE chlorofluorocarbon measurements, we calculate that the proportion of the emissions coming from the Northern Hemisphere is between 73 and 81%, depending on the OH distribution used. However this result includes an adjustment of 5% derived from a simulation of the 2D estimation procedure using the 3D MOZART model. This adjustment is needed because of the very different spatial emission distributions of the chlorofluorocarbons and methane which makes chlorofluorocarbon derived transport rates

inaccurate for the 2D simulation of methane. The 2D model combined with the annual cycle in OH from Spivakovsky *et al.* (2000) provides an acceptable fit to the observed 12 month cycles in methane. The trend in the amplitude of the annual cycle of methane at Cape Grim is used to infer a trend in OH in 30-90°S of 0% ± 5%/decade from 1985 to 2000, in qualitative agreement with Prinn *et al.* (2001) for the Southern Hemisphere.

Continuous high frequency observations of hydrogen at the Mace Head baseline atmospheric monitoring station over the 1994-1998 period, *J. Geophys. Res.*, 2000

P. G. Simmonds, R.G. Derwent, S. O'Doherty, D.B. Ryall, L.P. Steele, R. L. Langenfelds, P. Salameh, H. J. Wang, C.H. Dimmer, and L.E. Hudson.

Abstract. Continuous high frequency (every 40 minutes) automatic measurements of hydrogen have been made at the Mace Head atmospheric research station on the Atlantic Ocean coast of Ireland throughout 1994-1998. These observations represent one of the most comprehensive *in-situ* records of a trace gas that has received comparatively little attention. Individual measurements have been sorted by four independent methods to separate clean, maritime air masses from regionally polluted European air masses. Hydrogen concentrations in mid-latitude Northern Hemisphere baseline air show a distinct seasonal cycle with highest concentrations during spring and lowest concentrations during late autumn, with a peak-to-trough amplitude of 38 ± 6 ppb, averaged over the observed seasonal cycles from 1994 to 1998. The mean hydrogen concentration in mid-latitude northern hemisphere baseline air on 1st January 1995 was estimated as 496.5 ppb with an upwards trend of 1.2 ± 0.8 ppb yr^{-1}. Evidence has also been obtained for European pollution sources with source strength of about 0.8 Tg yr^{-1} and for deposition of hydrogen to soils. The observation of slightly elevated hydrogen concentrations relative to baseline levels in tropical maritime air masses points to a latitudinal gradient in hydrogen with higher concentrations in lower latitudes of the Northern Hemisphere and in the Southern Hemisphere. This is confirmed by comparable hydrogen observations at Cape Grim, Tasmania, which are consistently higher than measurements recorded at Mace Head. Mean hemispheric concentrations of 504 and 520 ppb have been estimated for the northern and southern hemispheres, respectively, for 1st January 1996, corresponding to a total atmospheric hydrogen burden of 182 Tg.

In situ chloroform measurements at Advanced Global Atmospheric Gases Experiment atmospheric research stations from 1994 to 1998, *J. Geophys. Res.*, 2001

S. O'Doherty, P.G. Simmonds, D. M. Cunnold, H. J. Wang, G. A. Sturrock, P. J. Fraser, D. Ryall, R. G. Derwent, R. F. Weiss, P. Salameh, B. R. Miller, and R. G. Prinn

Abstract. Measurements of atmospheric chloroform ($CHCl_3$) by in situ gas chromatography using electron capture detection are reported from the Advanced Global Atmospheric Gases Experiment (AGAGE) network of atmospheric research stations. They are some of the most comprehensive *in situ*, high-frequency measurements to be reported for $CHCl_3$ and provide valuable information not only on clean "baseline" mixing ratios but also on local and regional sources. Emissions from these sources cause substantial periodic increases in $CHCl_3$ concentrations above their baseline levels, which can be used to identify source strengths. This is particularly the case for measurements made at Mace Head, Ireland. Furthermore, these local sources of $CHCl_3$ emissions are significant in relation to current estimates of global emissions and illustrate that the understanding of competing sources and sinks of $CHCl_3$ is still fragmentary. These observations also show that $CHCl_3$ has a very pronounced seasonal cycle with a summer minimum and winter maximum presumably resulting from enhanced destruction by OH in the summer. The amplitude of the cycle is dependent on sampling location. Over the 57 months of *in situ* measurements a global average baseline concentration of 8.9±0.1 ppt was determined with no appreciable trend in the baseline detected.

European greenhouse gas emissions estimated from continuous atmospheric measurements and radon 222 at Mace Head, Ireland, *J. Geophys. Res.*, 2000

S. Biraud, P. Ciais, M. Ramonet, P. Simmonds, V. Kazan, P. Monfray, S. O'Doherty, T. G. Spain, and S. G. Jennings

Abstract. Flux estimates of CO_2, CH_4, N_2O, and CFCs over western Europe have been inferred from continuous atmospheric records of these species at the atmospheric research station of Mace Head, Ireland. We use radon (^{222}Rn) which has a fairly uniform source over continents as a reference compound to estimate unknown sources of other species. The correlation between each species and ^{222}Rn is calculated for a suite of synoptic events that have been selected in the Mace Head record over the period 1996/97. In the following, we describe the method and its uncertainties, and we establish data selection criteria that minimize the influence of local sources over Ireland, in the vicinity of the station, in order to select synoptic events originating from western Europe. We estimate western European flux densities of 45-30 x 10^3 kg C km^{-2} $month^{-1}$ during wintertime for CO_2, of 4.8-3.5 x 10^3 kg CH_4 km^{-2} yr^{-1}, 475-330 kg N_2O km^{-2} yr^{-1}, 2.5-1.8 kg CFC-11 km^{-2} yr^{-1} for CFC-11, and 4.2-2.9 kg CFC-12 km^{-2} yr^{-1} for CFC-12. Our estimates are independent, although in good agreement with those produced by inventories, except for CFC-11 where our estimate is much lower than the inventory.

Estimating source regions of European emissions of trace gases from observations at Mace Head, *Atmos. Environ.*, 2001

D. B. Ryall, R. G. Derwent, A. J. Manning, P. G. Simmonds and S. O'Doherty

Abstract. A technique is described for identifying probable source locations for a range of greenhouse and ozone-depleting trace gases from the long term measurements made at Mace Head, Ireland. The Met. Office's dispersion model NAME is used to predict concentrations at Mace Head from all possible sources in Europe, then source regions identified as those which consistently lead to elevated concentrations at Mace Head. Estimates of European emissions and their distribution are presented for a number of trace gases for the period 1995-1998. Estimated emission patterns are realistic, given the nature and varied applications of the species considered. The results indicate that whilst there are limitations, useful information about source distribution can be extracted from continuous measurements at a remote site. It is probable that much improved estimates could be derived if observations were available from a number of sites. The ability to assess emissions has obvious implications in monitoring compliance with internationally agreed quota and protocols.

(2) MAJOR PUBLICATIONS

The central accomplishments of the ALE/GAGE/AGAGE program from 1999 to 2003 are documented in the following journal publications and theses. Other relevant 1999-2003 publications are given in Section (5).

Biraud, S., P. Ciais, M. Ramonet, P. Simmonds, V. Kazan, P. Monfray, and S. O'Doherty, European greenhouse gas emissions estimated from continuous atmospheric measurements and Radon 222 at Mace Head, Ireland, *J. Geophys. Res.*, **105**, 1351-1366, 2000.

Biraud, S., P. Ciais, M. Ramonet, P. Simmonds, V. Kazan, P. Monfray, S. O'Doherty, T. G. Spain, and S.G. Jennings, Quantification of Carbon Dioxide, Methane, Nitrous oxide, and Chloroform emissions over Ireland from atmospheric observations at Mace Head, *Tellus., 54B*, 41-60, 2002.

Cohan, D. S., G. A. Sturrock, A. P. Biazar, M. L. Cox and P. J. Fraser, Methyl iodide at Cape Grim, Tasmania from AGAGE observations, *J. Atmospheric Chemistry*, in press, 2002.

Cox, M. L., A regional study of the natural and anthropogenic sources and sinks of the major halomethanes, *PhD thesis*, School of Mathematical Sciences, Monash University, Clayton, Victoria, Australia, 2001.

Cox, M. L., G. A. Sturrock, P. J. Fraser, S. Siems, P. B. Krummel and S. O'Doherty, Regional sources of methyl chloride, chloroform and dichloromethane identified from AGAGE observations at Cape Grim, Tasmania, 1998-2000, *J. Atmospheric Chemistry*, in press, 2002.

Cox, M. L., G. A. Sturrock, S. Siems and P. J. Fraser, TAPM modelling studies of AGAGE dichloromethane observations at Cape Grim, *Baseline 1999-2000*, in press, 2002.

Cox, M., P. Hurley, P. J. Fraser and W. Physick, Investigation of Melbourne region pollution events using Cape Grim AGAGE carbon monoxide data, a regional transport model (TAPM) and the EPA Victoria carbon monoxide inventory, *Clean Air*, **33**, 35-40, 2000.

Cunnold, D., L. P. Steele, P. J. Fraser, P. G. Simmonds, R. G. Prinn, R. F. Weiss, L. W. Porter, S. O'Doherty, R. L. Langenfelds, P. B. Krummel, H. J. Wang, L. Emmons, X. X, Tie and E. J. Dlugokencky, *In situ* measurements of atmospheric methane at GAGE/AGAGE sites during 1985 to 2000 and resulting source inferences, *J. Geophysical Research*, in press, 2002.

Derwent, R.G., D.B. Ryall, S.G. Jennings, T.G. Spain, and P. G. Simmonds, Black Carbon Aerosol and Carbon Monoxide in European Regionally-Polluted Air Masses at Mace Head, Ireland During 1995-1998, *Atmos. Environ.*, **35**, 6371-6378, 2001.

Derwent, R.G., N. Carslaw, P. G. Simmonds, M. Bassford, S. O'Doherty, D.B. Ryall, M.J. Pilling, A.C. Lewis, and J.B. McQuaid, Hydroxyl radical concentrations estimated from measurements of trichloroethylene during the EASE/ACSOE campaign at Mace Head, Ireland during July 1996, *J. Atmos. Chem.*, **34**, 185-205, 1999.

Dimmer, C.H., Sources, Abundances and Seasonality of Tropospheric Halocarbons in the remote Northern Hemisphere, *Ph.D Thesis*, University of Bristol, Bristol, United Kingdom, 412 pp., 1999.

Dunse, B. L., Investigation of urban emissions of trace gases by use of atmospheric measurements and a high-resolution atmospheric transport model, *PhD thesis*, Department of Chemistry, University of Wollongong, Wollongong, New South Wales, Australia, submitted, 2002.

Dunse, B. L., L. P. Steele, P. J. Fraser and S. R. Wilson, An analysis of Melbourne pollution episodes observed at Cape Grim from 1995-1998, *Baseline 1997-1998*, 34-42, 2001.

Greally, B.R., Development of an analytical system for the determination of highly fluorinated compounds in air samples, *Ph.D Thesis*, University of Bristol, Bristol, United Kingdom, 363 pp., 1999.

Huang, J., Optimal determination of global hydroxyl concentrations using multiple trace species, *PhD thesis, Report No. 65*, Center for Global Change Science, Department of Earth, Atmosphere and Planetary Sciences, MIT, 2000.

Huang, J. and R. G. Prinn, Critical evaluation of emissions for potential new OH estimation gases, *J. Geophysical Research*, in press, 2002.

Jensen, C. D., Terrestrial sources and sinks of atmospheric methyl bromide: three-dimensional modelling of tropospheric abundance and sensitivities, *MSc thesis, Report No. 62*, Center for Global Change Science, Department of Earth, Atmosphere and Planetary Sciences, MIT, 1999.

Manning A.J., D.B. Ryall, R.G. Derwent, P.G. Simmonds, and S.O'Doherty, Estimating European Emissions of ozone-depleting and greenhouse gases using observations and a Modelling Back-Attribution Technique, *Atmos. Environ.*, submitted, 2002.

McCulloch A., P. Ashford and P.M. Midgley, Historic emissions of fluorotrichloromethane (CFC-11) based on a market survey, *Atmos. Environ.*, **35***(26)*, 4387-4397, 2001.

McCulloch A. and P.M. Midgley, The history of methyl chloroform emissions: 1951 - 2000, *Atmos. Environ.*, **35***(31)*, 5311-5319, 2001.

Midgley P.M. and A. McCulloch, *Production, Sales and Emissions of Halocarbons from Industrial Sources*, Ch. 6 of The Handbook of Environmental Chemistry Vol 4. Part E,

Reactive Halogen Compounds in the Atmosphere (ed. P. Fabian and O.N. Singh), Springer-Verlag, Heidelberg, 1999.

O'Doherty, S., P. Simmonds, D. Cunnold, H. J.Wang, G.A. Sturrock, P.J. Fraser, D. Ryall, R.G. Derwent, R.F. Weiss, P. Salameh, B.R. Miller, and R. G. Prinn, In-situ chloroform measurements at Advanced Global Atmospheric Gases Experiment atmospheric research stations from 1994-1998, *J. Geophys. Res.*, **106**, 20,429-20,444, 2001.

Prinn, R.G. and J. Huang, Comment on "Global OH trend inferred from methyl chloroform measurements" by M. Krol *et al.* (1998), *J. Geophysical Research*, **106**, 23151-23158, 2001.

Prinn, R.G, J. Huang, R. Weiss, D. Cunnold, P. Fraser, P. Simmonds, C. Harth, P. Salameh, S. O'Doherty, R. Wang, L. Porter, B. Miller and A. McCulloch, Evidence for significant variations of atmospheric hydroxyl radicals in the last two decades, *Science*, **292**, 1882-1888, 2001. Correction, *Science*, **293**, 1054, 2001.

Prinn, R. G., R. F. Weiss, P. J. Fraser, P. G. Simmonds, D. M. Cunnold, F. N. Alyea, S. O'Doherty, P. Salameh, B. R. Miller, J. Huang, R. H. J. Wang, D. E. Hartley, C. Harth, L. P. Steele, G. Sturrock, P. M. Midgley and A. McCulloch, A history of chemically and radiatively important gases in air deduced from ALE/GAGE/AGAGE, *Journal of Geophysical Research*, **105**, 17,751-17,792, 2000.

Ryall, D.B., R.G. Derwent, A.J. Manning, P.G. Simmonds, and S.O'Doherty, Estimating source regions of European emissions of trace gases from observations at Mace Head, *Atmos. Environ.*, **35**, 2507-2523, 2001.

Simmonds, P.G., R.G. Derwent, S. O'Doherty, D.B. Ryall, L.P. Steele, R.L. Langenfelds, P. Salameh, H.C. Wang, C., H. Dimmer, and L.E. Hudson, Continuous high-frequency observations of hydrogen at the Mace Head baseline atmospheric monitoring station over the 1994-1998 period, *J. Geophys. Res.*, **105**, 12,105-12,121, 2000.

Sturrock, G. A., D. M. Etheridge, C. M. Trudinger, P. J. Fraser and A. M. Smith, Reconstruction of atmospheric histories of halocarbons from analysis of Antarctic firn: major Montreal Protocol species, *J. Geophysical Research*, in press, 2002.

Sturrock, G. A., L. W. Porter and P. J. Fraser, *In situ* measurement of CFC replacement chemicals and other halocarbons at Cape Grim: the AGAGE GC-MS program, *Baseline 1997-1998*, 43-49, 2001.

Wang, Y.-P. and S.T. Bentley, Development of a spatially explicit inventory of methane emissions from Australia and its verification using atmospheric concentration data, *Atmospheric Environment*, in press, 2002.

(3) INTERNATIONAL ASSESSMENTS (1999-2003)

ALE/GAGE/AGAGE measurements and derived lifetimes, OH concentrations, and emissions are of considerable policy significance and are widely used in international ozone layer and climate assessments. In the 1999-2003 time frame, AGAGE team members have specifically contributed to the following major international assessments:

Kurylo, M. J., J. M. Rodriguez, M. O. Andreae, E. L. Atlas, D. R. Blake, J. H. Butler, S. Lal, D. J. Lary, P. M. Midgley, S. A. Montzka, P. C. Novelli, C. E. Reeves, P. G. Simmonds, L. P. Steele, W. T. Sturges, R. F. Weiss and Y. Yokouchi, Short-Lived Ozone-Related Compounds. Chapter 2 in: *Scientific Assessment of Ozone Depletion: 1998*. World Meteorological Organization, Global Ozone Research and Monitoring Project, Report 44, Geneva, 56 pp., 1999.

McCulloch, A., Contributing Author and Reviewer, Intergovernmental Panel on Climate Change, *Good Practice Guidance and Uncertainty Management in National Greenhouse Gas Inventories*, 2000.

McCulloch, A., D. Cunnold, P. Midgley and R. Weiss, Contributing Authors and Reviewers, Ch. 1 and 2, *Scientific Assessment of Ozone Depletion: 2002*, UNEP/WMO, in press, 2002.

McCulloch, A., and Prinn, R.G., Intergovernmental Panel on Climate Change (IPCC), Reviewers of Working Group I Report, *Climate Change 2001: The Scientific Basis,* Cambridge U. Press, 2001.

Midgley, P., Intergovernmental Panel on Climate Change (IPCC), Lead Author in Working Group I Report, 2001.

Montzka, S. and P. Fraser, Lead Authors, Controlled Substances and Other Source Gases, Chapter 1 in: *Scientific Assessment of Ozone Depletion: 2002*, UNEP/WMO, in press, 2002.

Prather, M. and D. Ehhalt, Coordinating Lead Authors, Atmospheric Chemistry and Greenhouse Gases, Chapter 4 in: *Climate Change 2001: the Scientific Basis*, J. T. Houghton *et al.* (eds.), Cambridge University Press, 239-287, 2001.

Prinn, R. G., R. Zander, D. M. Cunnold, J. W. Elkins, A. Engel, P. J. Fraser, M. R. Gunson, M. K. W. Ko, E. Mahieu, P. M. Midgley, J. M. Russell III, C. M. Volk and R. F. Weiss, Long-Lived Ozone-Related Compounds, Chapter 1 in: *Scientific Assessment of Ozone Depletion: 1998*, World Meteorological Organization, Global Ozone Research and Monitoring Project, Report 44, Geneva, 54 pp., 1999.

Van Roozendael, M. and G. Vaughan, Long-Term Changes, Chapter 2 in: *European Research in the Stratosphere*, Directorate-General for Research, European Commission, Brussels, Belgium, 29-67, 2001.

(4) NATIONAL ASSESSMENTS (1999-2003)

AGAGE data and/or AGAGE scientists have also contributed to various national assessments.

The UK government uses the Mace Head data in its official emission inventory validation for greenhouse gases. Details are available in the following report: Verification of the UK estimates of the Kyoto gases, R. Derwent, Appendix 10, In: *UK Greenhouse Gas Inventory, 1990 to 2000.* Annual report for submission under the Framework Convention on Climate Change, A.G. Salway, T.P. Murrells, R. Milne, and S Ellis. NETCEN Report ISBN 0-7058-1805-5, Culham, Oxfordshire, UK, March 2002.

The Mace Head, Ireland observations coupled with the UK Meteorological Office particle dispersion "NAME" model have also been used to provide greenhouse gas inventories for the Irish Government.

The following papers use AGAGE Cape Grim data and/or involve AGAGE scientists to assess Australian and New Zealand environmental issues:

ANZECC, *Core Environmental Indicators for Reporting on the State of the Environment*, Australian and New Zealand Environment and Conservation Council, State of the Environment Reporting Task Force, Environment Australia, Canberra, Australian Capital Territory, Australia, 92 pp, 2000.

Bek, P., P. Crosser, H. Hofman, P. Latch and D. Traynor, *State of the Environment Queensland 1999*, Queensland Government EPA, Brisbane, Australia, 1999.

Manins, P., P. Holper, R. Suppiah, R. Allan, K. Walsh, P. Fraser and T. Beer, Atmosphere, Chapter 1 in: *Australia State of the Environment 2001*, Department of Environment and Heritage/CSIRO, Canberra, Australian Capital Territory, Australia, 145 pp, 2001.

(5) OTHER RELEVANT PUBLICATIONS (1999-2003)

Here we list other publications in the 1999-2003 time period supported by, and/or relevant to, AGAGE. These include technique papers, program reports, meeting abstracts, and advanced theoretical analyses of AGAGE data:

Alexander, B., M. K. Vollmer, T. Jackson, R. F. Weiss and M. H. Thiemens, Stratospheric CO_2 isotopic anomalies and SF_6 and CFC tracer concentrations in the Arctic polar vortex, *Geophysical Research Letters*, **28**, 4103-4106, 2001, and; Correction to "Stratospheric CO_2 isotopic anomalies and SF_6 and CFC tracer concentrations in the Arctic polar vortex" *Geophysical Research Letters*, **29**, No. 5, 10.1029/2002GL014749, 2002.

Bai, J., M. Wang, J. Graham, R.G. Prinn, G. Kong, and Z. Huang, The analysis for the variation characteristics of surface ozone and NO_x in Dinghushan station, *Acta Scientiae Circumstantiae*, **19**, 262-265, 1999.

Bai, J., M. Wang, J. Graham, R.G. Prinn, Z. Huang and G. Kong, The study of the relationships of surface ozone, NO_x and solar visible radiation in Dinghushan station, *Acta Scientiae Circumstantiae*, **20**, 173-178, 2000.

Bassford, M.R., G. Nickless, P. Simmonds, A.C. Lewis, M.J. Pilling, M.J. Evans and J.A. Pyle, Concurrent observations of alkyl halides and dimethyl sulphide in marine air: Implications for sources of atmospheric methyl iodide. *Atmos. Environ.*, **33**, 2373-2383 (1999).

Corno, G., A. McMinn, G. Sturrock, R. Parr, N. Tindale, L. Porter, R. Gillett, P. Fraser, C. Reeves and S. Penkett, A preliminary investigation of the phytoplankton ecology and biogenic gas production of Cape Grim, NW Tasmania, *Baseline 1999-2000*, in press.

Cunnold, D. M. and R. G. Derwent, Emission estimates from ground-based measurements, *Proceedings of the International Workshop on Emissions of Chemical Species and Aerosols into the Atmosphere*, (POET, NILU, Norway) in press, 2002.

Derwent, R.G., D.B. Ryall, A.J. Manning, P.G. Simmonds, S. O'Doherty, S. Biraud, P. Ciais, M. Ramonet, and S.G. Jennings, Continuous observations of carbon dioxide at Mace Head, Ireland from 1995-1999 and its net European ecosystem exchange, *Atmos. Environ.*, **36**, 2799-2807, 2002.

Dimmer, C.H., P.G. Simmonds, G.N. Nickless, and M. R. Bassford, Biogenic fluxes of halomethanes from Irish Peatland Ecosystems, *Atmos. Environ.*, , 321-330, 2000.

Dimmer, C.H., A. McCulloch, and P. Simmonds, Tropospheric concentrations of the chlorinated solvents, tetrachloroethene, trichloroethene, measured in the remote Northern Hemisphere, *Atmos Environ.*, , 1171-1182, 2001.

Enting, I.G., Inverse Problems in Atmospheric Constituent Transport, Cambridge U. Press, Cambridge, 392 pgs., 2002.

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