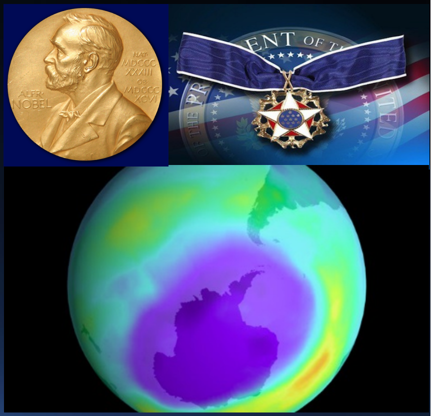
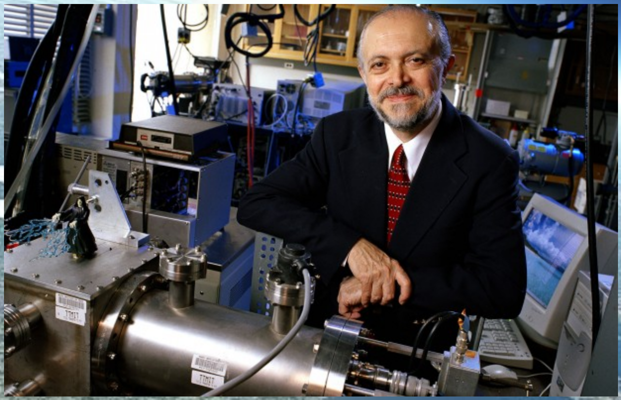
Mario J. Molina Symposium

Atmospheric Chemistry, Climate, and Policy





The Symposium recognizes Dr. Molina's pioneer research of the impacts of chlorofluorocarbons (CFCs) on stratospheric ozone that demonstrated the vulnerability of the natural environment and promoted the public awareness of the potential harmful consequences of anthropogenic activities. Sponsors include the American Meteorological Society, National Science Foundation, Department of Energy, National Aeronautics and Space Administration, World Meteorological Organization, and International Commission on Atmospheric Chemistry and Global Pollution

AMS
American Meteorological Society

96th
Annual
Meeting

10-14
January
2016

THE 2016 AMS MARIO J. MOLINA SYMPOSIUM

FOREWORD

Dr. Molina first demonstrated that industrially produced chlorofluorocarbons (CFCs) decompose in the stratosphere and release chlorine atoms, leading to catalytic ozone destruction. His research in stratospheric chemistry was instrumental to the establishment of the 1987 United Nations Montreal Protocol to ban ozone-depleting substances worldwide. Dr. Molina's contributions to preserving the planet Earth not only save the atmospheric ozone layer, but also protect the climate by reducing the emissions of greenhouse gases. He was awarded the 1995 Nobel Prize in Chemistry for his pioneering research in understanding the stratospheric ozone loss mechanism. In 2013, President Barack Obama announced Dr. Molina as a recipient of the Presidential Medal of Freedom.

The 2016 AMS Molina Symposium honors Dr. Molina's distinguished contributions to research related to atmospheric chemistry. The symposium contains an integrated theme related to atmospheric chemistry, climate, and policy.

EDUCATIONAL BACKGROUND

1965, B.A. Chemical Engineering, National Autonomous University of Mexico
1972, Ph.D., Chemistry, University of California, Berkeley

PROFESSIONAL POSITIONS

Assistant Professor, Universidad Nacional Autónoma de México, 1967-68
Postdoctoral Associate, University of California, Berkeley, 1972-73
Postdoctoral Associate, University of California, Irvine, 1973-75
Assistant Professor, University of California, Irvine, 1975-79
Associate Professor, University of California, Irvine, 1979-82
Member of Technical Staff, Jet Propulsion Laboratory, Caltech, 1982-84
Senior Research Scientist, Jet Propulsion Laboratory, Caltech, 1984-89
Professor, Dept. of Earth, Atmospheric & Planetary Sciences and Dept. of Chemistry; Lee and Geraldine Martin Professor of Environmental Studies (1992-97); Institute Professor (1997), Massachusetts Institute of Technology (1989-2003)
President, Mario Molina Center for Strategic Studies in Energy and Environment, 2003-present
Professor, Dept. of Chemistry and Biochemistry, and Scripps Institution of Oceanography, University of California, San Diego, 2003-present

HONORS AND AWARDS

UCI Alumni Association Special Recognition for Contributions in Basic Research, 1976
Newport Democratic Club Public Service Award for Service to the Environment, 1977
Alfred P. Sloan Foundation Fellow, 1976-78
Camille and Henry Dreyfus Teacher-Scholar, 1978-82
Tyler Ecology and Energy Prize, 1983
Society of Hispanic Professional Engineers Award for Achievement in Science and Technology, 1983
Council for Recognition of Hispanics, Science Honoree, 1984
American Chemical Society Esselen Award, 1987
American Association for the Advancement of Science Newcomb-Cleveland Prize, 1987-88
NASA Medal for Exceptional Scientific Achievement, 1989
United Nations Environment Program Global 500 Award, 1989
Orange County Section of the American Chemical Society Service through Chemistry Award, 1989
Pew Scholar on Conservation and the Environment, 1990-92

Member, National Academy of Sciences, 1993-present
Member, Academia Mexicana de Ingeniería, 1993-present
Max Planck Research Award, 1994-96
Nobel Prize in Chemistry, 1995
United Nations Environment Program Ozone Award, 1995
Doctor *Honoris Causa*, Universidad de Buenos Aires, Argentina, 1996
Walker Prize, Boston Museum of Science, 1996
Doctor *Honoris Causa*, Universidad Nacional Autónoma de México, México, 1996
Doctor Honoris Causa, University of East Anglia, Norwich, England, 1996
Golden Plate Award, American Academy of Achievement, 1996
Titular Member, European Academy of Arts, Sciences and Humanities, 1996
Member, Institute of Medicine, 1993-present
Member, Academia Mexicana de Ciencias, 1993-present
Honorary Member, American Meteorological Society, 1997
Associate Fellow, Third World Academy of Sciences, 1993-present
Honorary Degree, Doctor of Science, Yale University, 1997
Honorary Degree, Doctor of Laws, University of Calgary, Canada, 1997
American Chemical Society Award for Creative Advances in Environmental Science and Technology, 1998
American Geophysical Union Fellow, 1998
Doctor of Science, Honoris Causa, Connecticut College, New London, CT, 1998
Honorary Degree, Doctor of Science, Occidental College, Los Angeles, CA, 1998
Willard Gibbs Medal Award, 1998
American Physical Society Fellow, 1998
UNEP Sasakawa Prize, 1999
Member, Pontifical Academy of Sciences, 1993-present
Doctor Honoris Causa, Pontifica Universidad Católica del Perú, 2000
Doctor Honoris Causa, Universidad Nacional Mayor de San Marcos, Perú, 2000
Honorary Member, Sociedad Química del Perú, 2000
Doctor Honoris Causa, Universidad de las Américas, Puebla, Mexico, 2001
Honorary Degree, Doctor of Science, Trinity College, Connecticut, 2001
Honorary Degree, Doctor of Science, University of Miami, 2001
Fellow of the American Association for the Advancement of Science, 1993-present
Honorary Degree, Doctor of Science, University of Waterloo, Canada, 2002
Honorary Degree, Florida International University, 2002
Honorary Degree, Utah State University, 2002
Doctor Honoris Causa, Universidad de Pachuca, Mexico, 2002
Medalla al Mérito Ciudadano, Legislature of the Mexico City Government, 2002
Presea Ezequiel Montes Ledesma, Querétaro, Mexico, 2002
John P. McGovern Medal, Sigma Xi - The Scientific Research Society, 2002
Environment Award, Heinz Family Foundation, 2002
National Hispanic Scientist of the Year Award, MOSI, Tampa, Florida, 2003
Doctor of Science Honoris Causa, Tufts University, Massachusetts, 2003
Member, El Colegio Nacional, Mexico, 1993-present
Participant, International Council on Clean Transportation, 1993-present
Doctor Honoris Causa: Benemérita Universidad Autónoma de Puebla, Mexico, 2004
Doctor Honoris Causa, Universidad Autónoma Metropolitana, Mexico, 2004
Doctor of Science (Honorary Degree): University of South Florida, 2005
Doctor Honoris Causa, Universidad Autónoma del Estado de México, 2006
Doctor Honoris Causa, Universidad de Chile, 2006
Honorary Member, Society of Toxicology, 2007

Premio Nacional a la Excelencia Jaime Torres Bodet, Mexico, 2007
Premio Nacional Benito Juárez García al Mérito Ciudadano, Mexico, 2007
Doctor Honoris Causa, Colegio de Postgraduados (Institución de Enseñanza e Investigación en Ciencias Agrícolas), Mexico, 2007
Honorary Degree, The City College of New York, 2007
Máster de Oro, Spain, 2008
Gran Cruz de la Orden de Isabel la Católica, Spain, 2008
Doctor Honoris Causa, Universidad de Santiago de Chile, 2008
Presea Estado de México, "José María Luis Mora", Mexico, 2008
Honorary Member of Instituto Mexicano de Ingenieros Químicos, Mexico, 2008
Honorary Member of Fundación Carlos III, Spain, 2009
Doctor Honoris Causa: Centro de Investigación y de Estudios Avanzados del Instituto Politécnico Nacional, Mexico, 2009
Doctor Honoris Causa, Universidad Alfonso X El Sabio, Spain, 2009
Honorary Degree, Duke University, USA, 2009
Doctor Honoris Causa, Universidad de Guadalajara, Mexico, 2010
Doctor Honoris Causa, Univesité libre de Bruxelles, Belgium, 2010
Doctor Honoris Causa, Universidad del Valle de Mexico, Mexico, 2010
Doctor Honoris Causa, Universidad Nacional de San Luis Potosí, México, 2011
Officer In The Order Of Oranje-Nassau, Netherlands, 2011
Doctor Honoris Causa,Washington College, Maryland, 2011
Doctor Honoris Causa, University of British Columbia, Canada, 2011
Doctor Honoris Causa, Whittier College, California, 2012
Doctor of Science (Honorary Degree), Harvard University, Cambridge, Massachusetts, 2012
Doctor Honoris Causa, Universidad Complutense de Madrid, Spain, 2012
Doctor Honoris Causa, The University of Manchester, U.K., 2013
Medal "San Ignacio de Loyola", Universidad Iberoamericana, Mexico, 2013
Gold Medal of the President of the Italian Republic, 2013
Presidential Medal of Freedom, United States of America, 2013
Knight of the Legion of Honour, France, 2014
University of California San Diego Medal, 2014
Doctor of Science (Honorary Degree), John Jay College of Criminal Justice, New York, 2014
Honorary Doctor of Letters, Texas A&M University, 2015

OTHER PROFESSIONAL ACTIVITIES

Testified by invitation at hearings before the House of Representatives Subcommittee on Public Health & Environment of the Committee on Interstate and Foreign Commerce, 93rd Congress; the U.S. Senate Subcommittee on Toxic Substances and Environmental Oversight of the Committee on Environment and Public Works; and before numerous state legislatures on the issue of chlorofluorocarbons and stratospheric ozone depletion, 1974-1980
NSF Oversight Committee, Fluorocarbon Technology Assessment, Systems Control, Inc., 1976
NIH Advisory Committee to the Director, Recombinant DNA Research, 1977
NRC-NAS Commission on Human Resources, 1978-1981
NASA Panel for Chemical Kinetics and Photochemical Data Evaluation, 1978-2006
TRW/NASA Consultant, Halogen Occultation Experiment (HALOE) Langley Research Center, 1979-1981
Awards UCI Academic Senate, 1980-1981
NRC/NAS Panelist, Postdoctoral Fellowships Program, 1982-1987
Organizer and Presider, Symposium on Antarctic Ozone Depletion NCAR Scientific Programs Evaluation Committee, Panel Review of Atmospheric Chemistry Division, Boulder, CO, 1986
UCR Ad Hoc Committee for Review of the Statewide Air Pollution Research Center, Riverside, CA, 1988

AAAS Annual Meeting, Boston, MA 1988
NASA Advisory Committee to Oversee the Scientific Assessment of Atmospheric Effects Associated with Stratospheric Emissions from High Speed Civil Transports, 1988-1994
Editorial Advisory Board of the International Journal of Chemical Kinetics, 1988-1991
DOE Review Panel for the Molecular Science Research Center at the Pacific Northwest Laboratories, Batelle, Richland, WA, 1988
Organizer and Presider, Symposium on Physical Chemical Problems in the Earth's Atmosphere, 196th American Chemical Society National Meeting, Los Angeles, CA, 1988
Panelist, Annual Meeting of the National Academy of Sciences President's Circle on Issues Confronting the Global Future, Irvine, CA, 1989
NOAA Review Panel for the Aeronomy Laboratory (Chair), 1989
NSF Advisory Committee for Atmospheric Sciences, 1989-1994
NRC-NAS Steering Committee on Aeronautical Technologies, 1990-1993
Co-organizer and lecturer, Atmospheric Chemistry Workshop Universidad Nacional Autónoma de México, Mexico City, Mexico, 1991
Co-chair, Gordon Conference on Atmospheric Chemistry, New Hampton, NH, 1991
Co-chair, Atmospheric Chemistry Colloquium for Emerging Senior Scientists, MIT, Cambridge, MA, 1991
NRC-NAS Committee on Atmospheric Chemistry, 1991-1993
Sigma Xi National Lecturer, 1992-1993
Robert A. Welch Foundation Lecturer, 1994
NSF Advisory Committee for Geosciences, 1994-1997
President's Committee of Advisors on Science and Technology (PCAST), 1994-2000
Advisory Panel of Atmospheric Chemistry Division, National Center for Atmospheric Research, 1995-2003
Evaluation Committee for Chemistry Graduate Programs at the National Universidad Nacional Autónoma de México, Mexico City, Mexico, 1995
Member, MIT International Council, 1996-2000
Member, MIT Council on the Environment, 1996-2003
Member, MIT Task Force on Student Life and Learning, 1996-2000
Member, Editorial Advisory Board of Accounts of Chemical Research, 1996-2000
Member, Board of Directors, Northeast State Clean Air Foundation, 1996-2006
Advisor to the Camille and Henry Dreyfus Foundation (Environ. Program), 1996-2002
Chair, Advisory Board, United Nations Environment Programme Regional Office for North America (UNEP/RONA), 1996-1998
Member of the Jury, King Jaime I Prize on the Defense of Nature, Valencia, Spain, 1996-present
Member, Board of Governors, US-Mexico Foundation for Science, 1996-2003
Participant, Intel International Science and Engineering Fair, 1997
Co-Organizer, Symposium on "Atmospheric Chemistry: from Local to Global Pollution," Fifth Chemical Congress of North America, Cancun, Mexico, 1997
Panelist, White House Roundtable on Global Climate Change (Washington, DC), 1997
Member, Board of Directors, Union of Concerned Scientists, 1997-present
Member, Steering Committee, MIT Consortium on Global Environmental Challenges, 1997-2001
Member, The President's Committee on the National Medal of Science, 1997-2000
Member, National Research Council Board on Environmental Studies and Toxicology, 1997-2000
Member, Secretary of Energy Advisory Board, 1998-2002
Chair, Committee on Atmospheric Chemistry, National Academy of Sciences, 1999-2003
Member, Advisory Editorial Board, Chemical Physics Letters, 1999-2006
Chair, World Bank Blue Ribbon Panel on Mexico City Air Quality Management, 1999-2001
Co-Chair, PCAST Independent Review Board of the National Assessment on Climate Change, 1999-2000
Secretary of Energy Advisory Board Panel on Emerging Technologies, 2000
Member, Board of Trustees, Science Service, 2000-2005

Member, Visiting Committee, Max Planck Institute for Chemistry, 2000-2004
Member, National Research Council Board on Atmospheric Sciences and Climate, 2001-2004
Member, College of Chemistry Advisory Board, University of California, Berkeley, 2001-present
Chair, Environmental Sciences Review Committee, Brookhaven National Laboratory, 2002
Co-Chair, Workshop on the Environment, Challenges for the Chemical Sciences in the 21st Century, National Academy of Sciences, 2002
Member, Advisory Board, Journal of Physical Chemistry, 2002
Member, National Commission on Energy Policy, 2002-present
Chair, Scientific Advisory Council, Comisión Federal para la Protección contra Riesgos Sanitarios, Mexico, 2002-present
Member, Board of Directors, The John and Catherine MacArthur Foundation, 2002-present
Review Editor, Drafting Author of Summary for Policy Makers, Working Group I, Fourth Assessment Report of the Intergovernmental Panel on Climate Change, 2006-2007
Member, Advisory Council, ClimateWorks, 2008
Member, Advisory Council World Wildlife Fund, 2008
Member of The President's Council of Advisors on Science and Technology (PCAST), 2008-present
Board of Directors, Innovación en la Enseñanza de la Ciencia (INNOVEC) Foundation, 2008-present
Member of the External Advisory Board, MIT Energy Initiative, 2008-present
Member of the International Council on Clean Transportation, 2008-present
Testimony before the U.S. House of Representative, Hearing of the Committee on Energy Independence and Global Warming, 2010
Member of the InterAcademy Council Committee to Review the IPCC, 2010
Chair of the Science Advisory Council of the Stockholm Environmental Institute (SEI), 2012
Chair of the AAAS Climate Science Panel to the What We Know Initiative, 2014

FORMER STUDENTS/POST DOCS/RESEARCH SCIENTISTS/VISITING SCIENTISTS

Graduate Students

Keith Beyer; Keith Broekhuizen; John Edwards; Andres Flores; Kirsten Johnson; Judy Kim; Rose Koch; John Lamb; Dana Leard; Jennifer Lipson; Yves Mantz; James McMahon; V. Faye McNeill; Jong Ho Park; Dara Salcedo; John Seeley; Geoffrey Smith; Craig A. Smith; Darryl Spencer; Robert A. Stachnik; Karen Thundiyil; Samudra Vijay; Miguel Zavala; Renyi Zhang; Bilal Zuberi

Post-Docs/Research Scientists/Visiting Scientists

Jonathan Abbatt; Gustavo Arguello; Thomas W. Beiderhase; Allan Bertram; Anatoli Bogdan; Manjula Canagaratna; Telma Castro; David Dai; Edward Dunlea; Matthew Elrod; Steffen Frey; Benjamin De Foy; Yael Dubowski; Franz Geiger; Lukas Gutzwiller; Markus Haider; Martin Hunter; Takashi Ishiwata; Andrey Ivanov; John Jayne; Abha Kapilashrami; Ingrid Kohl; Thomas Koop; Shan-Hu Lee; Yong Liu; Thomas Loerting; Linsey Marr; Scot Martin; Roger Meads; Andre W Miziolek; Agustin Garcia Reynoso; Jun Noda; Carl Percival; Ulrich Pöschll; Phillip M. Sheehy; Gustavo Sosa; Francisco Tablas; Rodolfo Lacy Tamayo; Sofia Trakhtenberg; Rainer Volkamer; Jason West; Paul Wooldridge

THE ORGANIZING COMMITTEE

Renyi Zhang (chair), William Brune (co-chair), John P. Burrows, Jeff Collett, Jiwen Fan, Franz Geiger, Shan-Hu Lee, and Sasha Madronich.

SYMPOSIUM SPONSORS

Funding for the 2016 AMS symposium was provided by American Meteorological Society, National Science Foundation (NSF), Department of Energy (DOE), National Aeronautics and Space Administration (NASA), and World Meteorological Organization (WMO). The international Commission on Atmospheric Chemistry and Global Pollution (iCACGP) was a sponsor of this symposium.

MARIO J. MOLINA - BIOGRAPHICAL

I attended elementary school and high school in Mexico City. I was already fascinated by science before entering high school; I still remember my excitement when I first glanced at paramecia and amoebae through a rather primitive toy microscope. I then converted a bathroom, seldom used by the family, into a laboratory and spent hours playing with chemistry sets. With the help of an aunt, Esther Molina, who was a chemist, I continued with more challenging experiments along the lines of those carried out by freshman chemistry students in college. Keeping with our family tradition of sending their children abroad for a couple of years, and aware of my interest in chemistry, I was sent to a boarding school in Switzerland when I was 11 years old, on the assumption that German was an important language for a prospective chemist to learn. I remember I was thrilled to go to Europe, but then I was disappointed in that my European schoolmates had no more interest in science than my Mexican friends. I had already decided at that time to become a research chemist; earlier, I had seriously contemplated the possibility of pursuing a career in music - I used to play the violin in those days. In 1960, I enrolled in the chemical engineering program at UNAM, as this was then the closest way to become a physical chemist, taking math-oriented courses not available to chemistry majors.

After finishing my undergraduate studies in Mexico, I decided to obtain a Ph.D. degree in physical chemistry. This was not an easy task; although my training in chemical engineering was good, it was weak in mathematics, physics, as well as in various areas of basic physical chemistry - subjects such as quantum mechanics were totally alien to me in those days. At first I went to Germany and enrolled at the University of Freiburg. After spending nearly two years doing research in kinetics of polymerizations, I realized that I wanted to have time to study various basic subjects in order to broaden my background and to explore other research areas. Thus, I decided to seek admission to a graduate program in the United States. While pondering my future plans, I spent several months in Paris, where I was able to study mathematics on my own and I also had a wonderful time discussing all sorts of topics, ranging from politics, philosophy, to the arts, etc., with many good friends. Subsequently, I returned to Mexico as an Assistant Professor at the UNAM and I set up the first graduate program in chemical engineering. Finally, in 1968 I left for the University of California at Berkeley to pursue my graduate studies in physical chemistry.

During my first year at Berkeley, I took courses in physics and mathematics, in addition to the required courses in physical chemistry. I then joined the research group of Professor George C. Pimentel, with the goal of studying molecular dynamics using chemical lasers, which were discovered in his group a few years earlier.

George Pimentel was also a pioneer in the development of matrix isolation techniques, which is widely used in the study of the molecular structure and bonding of transient species. He was an excellent teacher and a wonderful mentor; his warmth, enthusiasm, and encouragement provided me with inspiration to pursue important scientific questions.

My graduate work involved the investigation of the distribution of internal energy in the products of chemical and photochemical reactions; chemical lasers were well suited as tools for such studies. At the beginning I had little experience with the experimental techniques required for my research, such as handling vacuum lines, infrared optics, electronic instrumentation, etc. I learned much of this from my colleague and friend Francisco Tablas, who was a postdoctoral fellow at that time. Eventually I became confident enough to generate original results on my own: my earliest achievement consisted of explaining some features in the laser signals - that at first sight appeared to be noise - as "relaxation oscillations," predictable from the fundamental equations of laser emission.

My years at Berkeley have been some of the best of my life. I arrived there just after the era of the free-speech movement. I had the opportunity to explore many areas and to engage in exciting scientific research in an intellectually stimulating environment. It was also during this time that I had my first experience dealing with the impact of science and technology on society. I remember that I was dismayed by the fact that high-power chemical lasers were being developed elsewhere as weapons; I wanted to be involved with research that was useful to society, but not for potentially harmful purposes.

After completing my Ph.D. degree in 1972, I stayed for another year at Berkeley to continue research on chemical dynamics. Then, in the fall of 1973, I joined the group of Professor F. Sherwood (Sherry) Rowland as a postdoctoral fellow, moving to Irvine, California. Sherry had pioneered research on "hot atom" chemistry, investigating chemical properties of atoms with excess translational energy and produced by radioactive processes. Sherry offered me a list of research options: the one project that intrigued me the most consisted of finding out the environmental fate of certain very inert industrial chemicals - the chlorofluorocarbons (CFCs) - which had been accumulating in the atmosphere and which at that time were thought to have no significant effects on the environment. This project offered me the opportunity to learn a new field - atmospheric chemistry - about which I knew very little; trying to solve a challenging problem appeared to be an excellent way to plunge into a new research area. The CFCs are compounds similar to others that Sherry and I had investigated from the point of view of molecular dynamics; we were familiar with their chemical properties, but not with their atmospheric chemistry.

Three months after I arrived at Irvine, Sherry and I developed the "CFC-ozone depletion theory." At first the research did not seem to be particularly interesting - I carried out a systematic search for processes that might destroy the CFCs in the lower atmosphere, but nothing appeared to affect them. We knew, however, that they would eventually drift to sufficiently high altitudes to be destroyed by solar radiation. The question was not only what destroys them, but more importantly, what the consequences are. We realized that the chlorine atoms produced by the decomposition of the CFCs would catalytically destroy ozone. We became fully aware of the seriousness of the problem when we compared the industrial amounts of CFCs to the amounts of nitrogen oxides which control ozone levels; the role of these catalysts of natural origin had been established a few years earlier by Paul Crutzen. We were alarmed at the possibility that the continued release of CFCs into the atmosphere would cause a significant depletion of the Earth's stratospheric ozone layer. Sherry and I decided to exchange information with the atmospheric sciences community: we went to Berkeley to confer with Professor Harold Johnston, whose work on the impact of the release of nitrogen oxides from the proposed supersonic transport (SST) aircraft on the stratospheric ozone layer was well known to us. Johnston informed us that months earlier Ralph Cicerone and Richard Stolarski had arrived at similar conclusions concerning the catalytic properties of chlorine atoms in the stratosphere, in connection with the release of hydrogen chloride either from volcanic eruptions or from the ammonium perchlorate fuel planned for the space shuttle.

We published our findings in Nature, in a paper which appeared in the June 28, 1974 issue. The years following the publication of our paper were hectic, as we had decided to communicate the CFC - ozone issue not only to other scientists, but also to policy makers and to the news media; we realized this was the only way to insure that society would take some measures to alleviate the problem.

To me, Sherry Rowland has always been a wonderful mentor and colleague. I cherish my years of association with him and my friendship with him and his wife, Joan. While he was on sabbatical leave in Vienna during the first six months of 1974, we communicated via mail and telephone. There were many exchanges of mail during this short period of time, which illustrated the frantic pace of our research at that time while we continued to refine our ozone depletion theory. Soon after, Sherry and I published several more articles on the CFC-ozone issue; we presented our results at scientific meetings and we also testified at legislative hearings on potential controls on CFCs emissions.

In 1975, I was appointed as a member of the faculty at the University of California, Irvine. Although I continued to collaborate with Sherry, as an assistant professor I had to prove that I was capable of conducting original research on my own. I thus set up an independent program to investigate chemical and spectroscopic properties of compounds of atmospheric importance, focusing on those that are unstable and difficult to handle in the laboratory, such as hypochlorous acid, chlorine nitrite, chlorine nitrate, peroxynitric acid, etc.

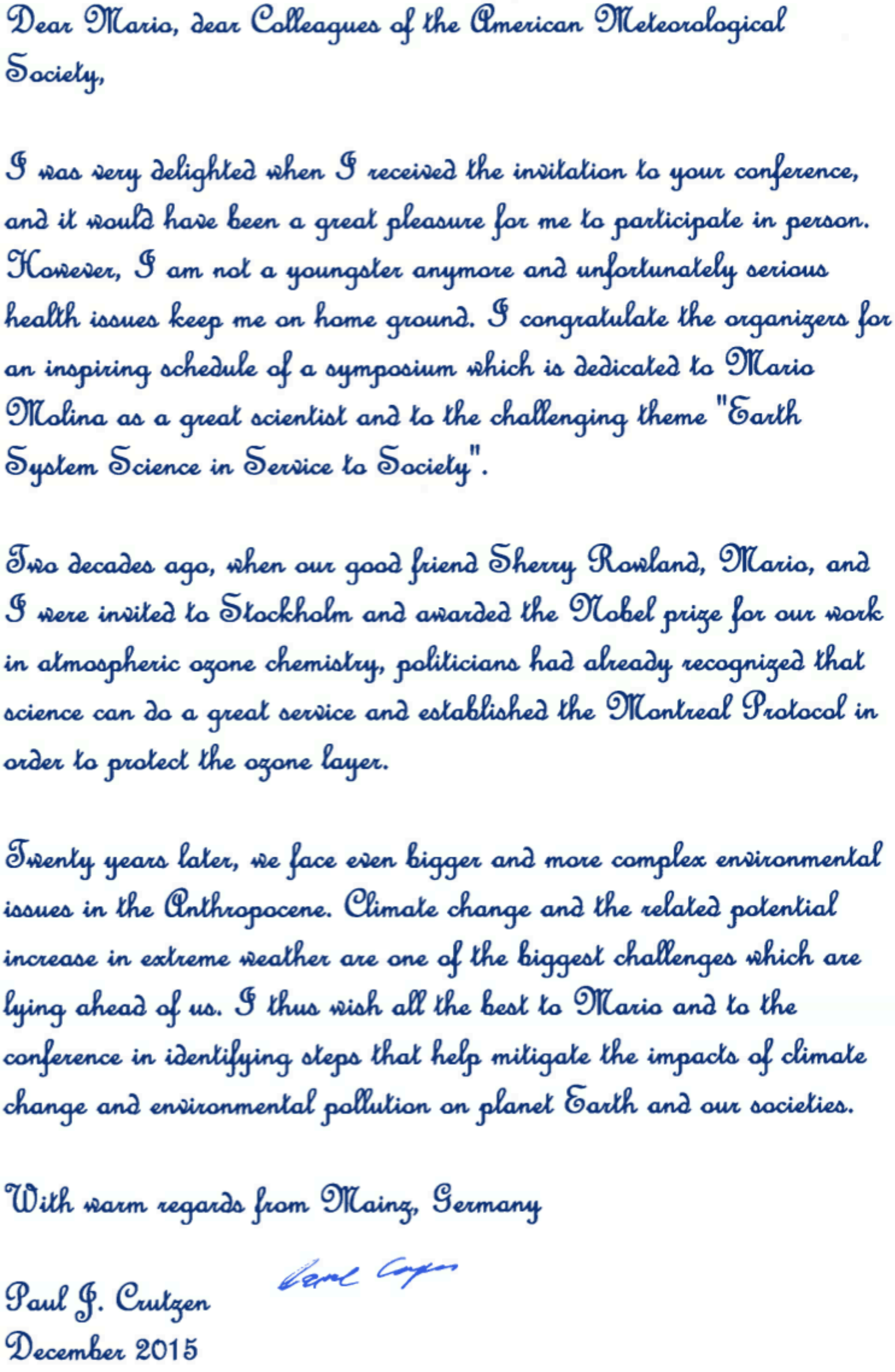
Although my years at Irvine were very productive, I missed not doing experiments myself because of the many responsibilities associated with a faculty position: teaching courses, supervising graduate students, meetings, etc. After spending seven years at Irvine as Assistant and then Associate Professor, I decided to move to a non-academic position. I joined the Molecular Physics and Chemistry Section at the Jet Propulsion Laboratory in 1982. I had a smaller group - only a few postdoctoral fellows - but I also had the luxury of conducting experiments with my own hands, which I enjoyed very much. Indeed, I spent many hours in the laboratory in those years, conducting measurements and developing techniques for the study of newly emerging problems. Around 1985, after becoming aware of the discovery by Joseph Farman and his co-workers of the seasonal depletion of ozone over Antarctica, my research group at JPL investigated the peculiar chemistry which is promoted by polar stratospheric clouds, some of which consist of ice crystals. We were able to show that chlorine-activation reactions take place very efficiently in the presence of ice under polar stratospheric conditions; thus, we provided a laboratory simulation of the chemical effects of clouds over the Antarctic. Also, in order to understand the rapid catalytic gas phase reactions that were taking place over the South Pole, experiments were carried out in my group with chlorine peroxide, a new compound which had not been reported previously in the literature and which turned out to be important in providing the explanation for the rapid loss of ozone in the polar stratosphere.

In 1989 I returned to academic life, moving to the Massachusetts Institute of Technology, where I have continued with research on global atmospheric chemistry issues.

Although I no longer spend much time in the laboratory, I very much enjoy working with my graduate and postdoctoral students, who provide me with invaluable intellectual stimulus. I have also benefited from teaching; as I try to explain my views to students with critical and open minds, I find myself continually being challenged to go back and rethink ideas. I now see teaching and research as complementary, mutually reinforcing activities.

When I first chose the project to investigate the fate of chlorofluorocarbons in the atmosphere, it was simply out of scientific curiosity. I did not consider at that time the environmental consequences of what Sherry and I had set out to study. I am heartened and humbled that I was able to do something that not only contributed to our understanding of atmospheric chemistry, but also had a profound impact on the global environment.

One of the very rewarding aspects of my work has been the interaction with a superb group of colleagues and friends in the atmospheric sciences community. I truly value these friendships, many of which go back 20 years or more, and which I expect to continue for many more years to come. I feel that this Nobel Prize represents recognition for the excellent work that has been done by my colleagues and friends in the atmospheric chemistry community on the stratospheric ozone depletion issue.



Dear Mario, dear Colleagues of the American Meteorological Society,

I was very delighted when I received the invitation to your conference, and it would have been a great pleasure for me to participate in person. However, I am not a youngster anymore and unfortunately serious health issues keep me on home ground. I congratulate the organizers for an inspiring schedule of a symposium which is dedicated to Mario Molina as a great scientist and to the challenging theme "Earth System Science in Service to Society".

Two decades ago, when our good friend Sherry Rowland, Mario, and I were invited to Stockholm and awarded the Nobel prize for our work in atmospheric ozone chemistry, politicians had already recognized that science can do a great service and established the Montreal Protocol in order to protect the ozone layer.

Twenty years later, we face even bigger and more complex environmental issues in the Anthropocene. Climate change and the related potential increase in extreme weather are one of the biggest challenges which are lying ahead of us. I thus wish all the best to Mario and to the conference in identifying steps that help mitigate the impacts of climate change and environmental pollution on planet Earth and our societies.

With warm regards from Mainz, Germany

Paul J. Crutzen
December 2015

“Scientist may depict problems that will affect the environment based on available evidence, but their solution is not the responsibility of scientists but of society as a whole” - Mario J. Molina

Dr. Molina and F.S. Rowland at University of California, Irvine, 1974

Ralph Cicerone, Paul Crutzen, Sherwood Rowland and Mario Molina, in Berlin, 1982.

The Nobel Prize in Chemistry 1995 was awarded jointly to Paul J. Crutzen, Mario J. Molina and F. Sherwood Rowland "for their work in atmospheric chemistry, particularly concerning the formation and decomposition of ozone". Press Conference / Nobel Prize -Stockholm, Sweden, December 10, 1995

Mario Molina receives Nobel Prize -Stockholm, Sweden, December 10, 1995

Signing of Stockholm Memorandum, Nobel Laureate Molina -Stockholm, Sweden, May 2011

Mario Molina receives from French President, François Hollande the Legion of Honor in degree of Knight, Mexico City, April 10, 2014

"Mario Molina is a visionary chemist and environmental scientist. Born in Mexico, Dr. Molina came to America to pursue his graduate degree. He later earned the Nobel Prize in Chemistry for discovering how chlorofluorocarbons deplete the ozone layer. Dr. Molina is a professor at the University of California, San Diego; Director of the Mario Molina Center for Energy and Environment; and a member of the President's Council of Advisors on Science and Technology."
Presidential Medal of Freedom Press Release, August 2013

Dr. Molina with President Bill Clinton - Washington, D.C., July 1997

President's Advisory Council on Science and Technology - PCAST November, 2013

Professor Molina with his students during an honorary symposium at UCSD in May 2014.

PROGRAM FOR MOLINA SYMPOSIUM BANQUET
JANUARY 12, 2016

Presiding: Misti L. Zamora, Texas A&M University; Franz M. Geiger, Northwestern University

7:30	Banquet starts - Room 238/239, New Orleans Ernest N. Morial Convention Center
7:30 – 7:40	Welcome Remarks by Dr. Alexander (Sandy) E. MacDonald, AMS President
8:40 – 8:45	Remarks by Paul J. Crutzen, Max Planck Institute for Chemistry
8:45 – 8:50	Remarks by Mark H. Thiemens, University of California San Diego
8:50 – 9:30	Open-floor Comments
9:30 – 9:40	Concluding Remarks by Mario J. Molina
9:40 – 9:45	Photo session
9:45	Adjourn

NOTES

J1.1

Real-time Mobile Lab Characterization of Urban Ambient Air Quality and Pollutant Emission Sources

C. E. Kolb, Aerodyne Research Inc., Billerica, MA; and T. I. Yacovitch, J. R. Roscioli, G. R. Magoon, and S. C. Herndon

Air quality in major urban and industrial regions is usually characterized by complex pollutant distributions that are often poorly captured by sparse fixed monitoring sites. In addition, accurate knowledge of primary air pollutant and secondary pollutant precursor emissions fluxes are necessary to predict pollutant concentration distributions and to determine which sources require mitigation to achieve and maintain healthy air quality. This challenge is particularly difficult in large urban and heavily industrialized areas with their diverse ranges and high densities of pollutant and pollutant precursor emission sources. Highly active atmospheric photochemistry leading to rapid evolution of both secondary gaseous pollutants and aerosol particulate matter poses additional challenges.

The use of suites of real-time trace gas and fine particle instruments (~1 s response times) deployed aboard mobile laboratories to map ambient pollutant distributions and detect, fingerprint, and quantify pollutant emission sources have proven to be valuable in large urban/industrial areas over the past ~15 years. One of the earliest demonstrations of mobile lab utility for major city air quality research occurred in 2002 and 2003 during a collaborative research program in the Mexico City Metropolitan Area (MCMA) led by Mario Molina's group at the Massachusetts Institute of Technology (MIT). This activity also included several Mexican and US academic and government research groups and featured mobile labs outfitted and operated by scientists from Aerodyne Research, Inc. (ARI). The Molina led MCMA-2002 and MCMA-2003 field campaigns in the largest megacity in North America served as a major test-bed to develop and demonstrate novel mobile lab pollutant measurement methods.

A large fraction of major air quality campaigns worldwide now feature mobile laboratory deployments to characterize ground level pollution distributions and sources. Recent evolution of both gaseous and fine particle pollutant real-time measurement capabilities and mobile lab measurement strategies will be illustrated with examples from recent field campaigns, including the 2015 Benzene and other Toxics Exposure (BEE-TEX) Study in Houston Texas.

J1.2

Globalizing the United States Air Quality Success Story

William Brune, Penn State University, University Park, PA

Air quality in the United States has improved dramatically in the past few decades. This success story is a result of government and industry response to the Clean Air Act, widespread monitoring of pollutants, improved understanding of photochemistry fundamentals, and economic decisions being made by industry, particularly the energy industry. Other developed countries have had their success stories as well. However, in much of the recently developed and developing world, the air quality is visually poor, usually poorly monitored, and unhealthy, causing millions of premature deaths annually. With current scientific understanding and technological know-how, the air quality could be improved in these countries, but there is an economic cost. In this talk, I will present some ideas on steps the United States could take to globalize the United States air quality success story.

J1.3

Reduction in Local Ozone Levels in Urban São Paulo Due to a Shift from Ethanol to Gasoline Use

Franz Geiger, Northwestern University, Evanston, IL

Ethanol-based vehicles are thought to generate less pollution than gasoline-based vehicles, because ethanol emissions contain lower concentrations of mono-nitrogen oxides than those from gasoline emissions. However, the predicted effect of various gasoline/ethanol blends on the concentration of atmospheric pollutants such as ozone varies between model and laboratory studies, including those that seek to simulate the same environmental conditions. Here, we report the consequences of a real-world shift in fuel use in the subtropical megacity of São Paulo, Brazil, brought on by large-scale fluctuations in the price of ethanol relative to gasoline between 2009 and 2011. We use highly spatially and temporally resolved observations of road traffic levels, meteorology and pollutant concentrations, together with a consumer demand model, to show that ambient ozone concentrations fell by about 20% as the share of bi-fuel vehicles burning gasoline rose from 14 to 76%. In contrast, nitric oxide and carbon monoxide concentrations increased. We caution that although gasoline use seems to lower ozone levels in the São Paulo metropolitan area relative to ethanol use, strategies to reduce ozone pollution require knowledge of the local chemistry and consideration of other pollutants, particularly fine particles. Nature Geoscience 7, 450–458 (2014) doi:10.1038/ngeo2144

J1.4

Elucidating Severe Urban Haze Formation in Beijing

Misti Levy Zamora, Texas A&M University, College Station, TX; and R. Zhang, S. Guo, and M. Hu

China has experienced severe haze pollution, with fine particulate matter concentrations reaching unprecedentedly high levels across many cities. During the fall of 2013, a suite of aerosol instruments was deployed in Beijing to directly measure a comprehensive set of aerosol properties, including the particle size distribution, hygroscopicity, and chemical composition. In this presentation, we will discuss the particulate matter formation mechanisms and potential mediation policies to minimize its regional to global impacts. We demonstrate that the periodic cycles of haze episodes in Beijing are largely driven by meteorological conditions and characterized by two distinct aerosol formation processes: nucleation and sustained particle growth. Nucleation consistently occurs during clean conditions, producing a high number concentration of nano particles. Accumulation of the particle mass concentration exceeding several hundred micrograms per cubic meter is attributed to a continuous size growth from the nucleation-mode particles over multiple days to yield numerous larger particles, distinctive from the aerosol formation typically observed in other regions worldwide. The particle compositions in Beijing exhibit a similarity to those commonly measured in other urban centers, which is indicative of chemical constituents dominated by secondary aerosol formation. Our results highlight that regulatory controls of gaseous emissions for volatile organic compounds and nitrogen oxides from local transportation and sulfur dioxide from regional industrial sources represent the key steps to reduce the urban PM level in China.

J3.1

Four Decades of Ozone Layer Depletion: What We Did Right and What We Could Have Done Different?

A. R. Ravishankara, Colorado State University, Fort Collins, CO

Molina and Rowland's seminal paper in 1974 led to the recognition of the global environmental problem of ozone layer depletion caused by anthropogenic emissions of chlorofluorocarbons. Subsequently, the list of substances that can deplete the ozone layer grew and included many chlorinated and brominated chemicals. The ozone layer depletion was evident on a global scale and dramatically in the Antarctic Spring time as the ozone hole. Faced with this global scale problem, amazingly, the nations of the world took action in adopting the Vienna Convention and subsequently the Montreal Protocol. Now, thanks to the seminal work of Molina and Rowland and the actions of the world community, the ozone layer is on the mend. Further more, the Protocol helped climate by stopping the production of the ozone depleting substances, which are also greenhouse gases.

Many things were done right in making Montreal Protocol a success. I will describe some of the features of the Montreal Protocol that enabled success. Then, we ask the question: Could something else been done using the chemicals and technologies that were already available prior to the Montreal Protocol or even subsequent to the Protocol? I will describe our work on potential pathways that could have been taken and what the ramifications of those steps would have been to the ozone layer and climate.

J3.2

An Historical Perspective on Ozone Research: From 1840 to 1970

Guy Brasseur, UCAR, Boulder, CO

Ozone was discovered in the laboratory by Schoenbein (1839) at the University of Basel, Switzerland. Laboratory studies by de LaRive and Marignac in Geneva (1845), by Becquerel and Frémy in France (1852), by Andrews in the UK (1856), and by Soret in Basel (1868) led to the identification of the chemical nature of ozone. The concept of antozone was introduced and later abandoned. Starting in the 1870's ozone was believed to provide a powerful therapy against several diseases. His presence in the atmosphere was detected for the first time by Houzeau in Rouen, France, and Albert Levy conducted systematic ozone measurements from 1877 to 1907 at Parc Montsouris in Paris. Extensive laboratory work was performed to investigate the absorption properties of ozone (Cornu, 1879; Hartley, 1881; Huggins, 1890; Fowler and Strutt, 1917). In 1920 Fabry and Buisson used these results to design a spectrograph, which became a prototype for the spectrophotometer developed by Dobson in Oxford, UK and is still used to date to accurately measure the ozone column. The first attempt to measure the vertical profile of ozone were made by Goetz (Spitzbergen, 1929), by Erich and Victor Regener (Germany, 1934) and from the manned balloon launched in 1935 in South Dakota. Theoretical progress in the understanding of the production and destruction processes resulted from the work of Chapman (1929), Bates and Nicolet (1950) and Hampson (1965). Interestingly, before the discovery of the ozone destruction mechanisms by chlorine (Stolarski, Cicerone, Rowland and Molina (USA, 1974), the formation of an "ozone hole" by injecting bromine and chlorine catalysts by rockets was considered in the early 1960's as a military action against a potential rival nation.

J3.3

The Future Ozone Layer Conundrum

Amy Hawes Butler, CIRES/Univ. of Colorado, Boulder, CO; and J. S. Daniel, D. W. Fahey, R. W. Portmann, A. R. Ravishankara, and K. H. Rosenlof

Future concentrations of carbon dioxide (CO2), nitrous oxide (N2O), and methane (CH4) will determine stratospheric ozone levels by 2100. We examine the range of possible ozone recovery with the Whole Atmosphere Community Climate Model by individually varying these three concentrations from strict mitigation scenarios to business as usual. If the increase in CO2 concentration slows down, then mitigating additional ODSs like N2O is important for avoiding further ozone depletion. However, if CO2 emissions continue as usual, reducing N2O will only exacerbate the extratropical ozone increase while marginally reducing tropical ozone depletion. We would thus face the conundrum of simultaneously protecting the tropics from increased ultraviolet (UV) exposure and the extratropics from reduced UV exposure. Future changes in methane can also have substantial consequences for ozone recovery.

J3.4

Ground-based Microwave Measurements of ClO in the Stratosphere

Gerald E. Nedoluha, Naval Research Laboratory, Washington, DC; and B. J. Connor, T. Mooney, J. W. Barrett, A. Parrish, R. M. Gomez, I. Boyd, D. R. Allen, M. Kotkamp, S. Kremser, T. Deshler, and M. L. Santee

We present 278 GHz ground-based microwave measurements of ClO in the lower stratosphere from Scott Base, Antarctica (since 1996), and in the upper stratosphere from Mauna Kea, Hawaii (since 1992). Both of these Chlorine Monoxide Experiment (ChlOE) instruments are a part of the Network for the Detection of Atmospheric Composition Change (NDACC). We use daytime and nighttime measurements together to form a day minus night spectrum, from which the difference in the day and night ClO profiles is retrieved. The ChlOE measurements at both locations are compared with ClO measurements from both UARS and Aura MLS.

ClO in the upper stratosphere is affected by variations in CH4 and by the solar cycle, hence seasonal and interannual variations are not necessarily representative of changes in chlorine, but since 1995 the ChlOE measurements near the peak of the ClO mixing ratio at Mauna Kea (20N) do show a decline in ClO of ~0.6%/year. Since the ChlOE measurements span the gap between the UARS and Aura MLS measurement periods they have also been used to show that, within the uncertainty of the ChlOE and MLS measurements, there is no significant bias between the two sets of MLS measurements.

At Scott Base (78S) we measure the large seasonal changes in lower stratospheric ClO which occur from mid-August through mid-October during the 'ozone hole' period, when temperatures are low enough for the formation of polar stratospheric clouds, and sunlight is available. The interannual variations in the measured ClO column over Scott Base are shown to be closely correlated with overall ozone mass depletion in September and October. They are also anti-correlated with the local interannual variations in 30 hPa temperatures from the MERRA reanalysis. Because of the complicated coupling of seasonal and diurnal variations of column ClO at these high latitudes a direct comparison of MLS and ChlOE measurements is difficult, but they do show similar sensitivities to interannual temperature variations. We make use a simple parameterization of the interannual 30 hPa temperature and column ClO variations in order to obtain chlorine trend estimates in this region.

J4.1

Anthropogenic Control of Biogenic SOA: Salt Interactions of Water Soluble Oxygenated Hydrocarbons

Rainer Volkamer, CIRES/Univ. of Colorado, Boulder, CO; and E. Waxman, T. Kurten, and J. Elm

Recent field observations suggest that aerosol sulfate (SO42-) directly controls a significant fraction of secondary organic aerosol (SOA) mass over biogenic environments such as the South Eastern US. Setschenow salting-constants, KS, describe the exponential dependence of the gas- / aqueous phase partitioning (Henry's Law constant) on the salt concentration. Knowledge about KS is of particular importance to predict SOA formation in aerosol water, where the salt concentrations are typically very high. Such salt-induced activity is not typically captured by activity coefficients, and can modify SOA formation rates by altering (1) the effective Henry's Law partitioning coefficient (by several orders of magnitude), (2) changing the multiphase chemical reaction rates; and (3) the volatity of products through the formation of complexes and/or organo-sulfate/nitrate products. This talk discusses laboratory measurements combined with quantum calculations to better understand the values of KS for biogenic OVOC, such as glyoxal, methyl glyoxal and isoprene epoxide in a range of atmospherically relevant salts, incl. (NH4)2SO4, NH4NO3, NaNO3 and NaCl. We find that glyoxal 'salts-in' (negative KS) consistently, while methyl glyoxal 'salts-out'. The Gibbs Energy of the chemical bonds of glyoxal, methyl glyoxal and isoprene epoxide with SO42-, NO3-, and Cl- is compared, and used to interrogate their salting-behavior. Our best estimates of KS values for glyoxal, methyl glyoxal and isoprene epoxide have been incorporated into the CMAQ model, and a range of simulations were conducted to predict the effect of salts on their partitioning and potential to form SOA over the continental US.

J4.2

Formation of High-Altitude Ice Clouds: Ice Nucleation or Freezing?

Anatoli Bogdan, Helsinki, Finland; and M. J. Molina

Upper tropospheric (UT) ice cirrus and type II ice PSCs are believed to be formed by freezing aqueous aerosol drops which contain up to 25–30 wt % of H2SO4 and HNO3. There are also reports that in UT aqueous drops, organic component may reach up to 50 wt %. Currently, atmospheric scientific community dealing with high-altitude ice clouds focuses only on the initial step of freezing, namely, on ice nucleation. How freezing proceeds after ice nucleation is not considered, although it is the freezing process itself which governs the phase state and surface properties of resulting cloud particles. Such limited consideration results in the lack of understanding of freezing process and, consequently, is a reason that some important problems of the formation and microphysics of UT cirrus and PSCs and the impact of these clouds on climate remain unsolved for decades. One of the important problems is whether freezing atmospheric aqueous drops produce completely solid ice particles, as is generally believed, or mixed-phased cloud particles in which an ice core is coated with a freeze-concentrated solution (FCS). About freeze-induced phase separation into pure ice and FCS1 and that mixed-phased cloud particles can be formed in UT2-5 and polar stratosphere6 have been reported some time ago. Unfortunately, atmospheric scientists dealing with UT cirrus and PSCs ignore these works that can stem from the fact that they cannot still comprehend and, consequently, accept the well-known fact of freeze-induced phase separation which occurs during the freezing of aqueous solutions, including atmospheric aqueous drops. The goal of this presentation is to present persuasive experimental results, including the visual demonstration of freezing process7, which would convince the atmospheric scientific community of the freeze-induced phase separation (Figure 1) and, consequently, of the formation of mixed-phase cloud particles at very beginning of UT cirrus and type II ice PSC development (Figure 1b). The knowledge of the phase state and surface properties of cloud particles (cloud microphysics) is important because they govern the rate of heterogeneous reactions destructing stratospheric and UT ozone and radiative properties of UT cirrus (absorption, reflection, and scattering of solar and terrestrial radiation). The FCS coating around UT cirrus particles strongly reduces the uptake of water vapor - the dominant greenhouse gas in UT - and, consequently, is responsible for the accumulation and persistence of elevated UT moisture.3,4 References: 1. Bogdan, A. Reversible formation of glassy water in slowly cooling diluted drops. J. Phys. Chem. B, 110, 12205-12206 (2006). 2. Bogdan, A., Molina, M. J., Sassen, K., Kulmala, M. Formation of low-temperature cirrus from H2SO4/H2O aerosol droplets. J. Phys. Chem. A, 110, 12541-12542 (2006). 3. Bogdan, A., Molina, M. J. Why does large relative humidity with respect to ice persist in cirrus ice clouds? J. Phys. Chem. A, 113, 14123-14130 (2009). 4. Bogdan, A., Molina, M. J. Aqueous aerosol may build up an elevated upper tropospheric ice supersaturation and form mixed-phase particles after freezing. J. Phys. Chem. A, 114, 2821-2829 (2010). 5. Bogdan, A., Molina, M. J. et. al. Solution coating around ice particles of incipient cirrus clouds. Proc. Natl. Acad. Sci. USA 11, E2439 (2013). 6. Bogdan, A., Molina, M. J. et. al. Formation of mixed-phase particles during the freezing of polar stratospheric ice clouds. Nature Chem., 2, 197-201 (2010). 7. Bogdan, A., Molina, M. J., et al. Visualization of freezing process in situ upon cooling and warming of aqueous solutions. Sci. Rep. 4, 7414 (2014).

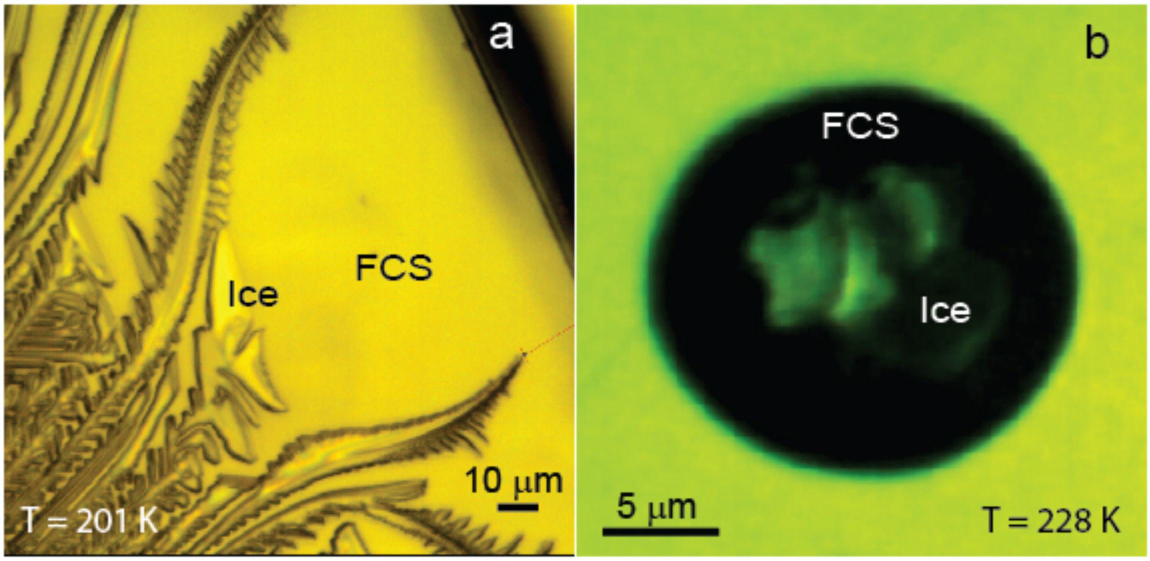


Figure 1. Images of frozen bulk 20 wt % CA (citric acid) (a) and finely dispersed 15 wt % H_2SO_4 drop (b).

J4.3

The Role of Coating-Surface Interactions in the Restructuring of Soot Nanoparticles

Alexei Khalizov, New Jersey Institute of Technology, Newark, NJ; and X. Fan

Atmospheric soot nanoparticles are fractal aggregates of graphitic spheres that are mixed with various organic and inorganic materials. The morphological and chemical complexity of soot nanoparticles is caused by diversity in emission sources and further amplified by aging transformations in the environment. The mechanistic details of these transformations, including the formation of specific mixing states and the extent and rate of morphological changes from the condensation of different materials remain poorly understood. Also, whereas it is commonly assumed that aging transformations produce compact and uniformly coated soot aggregates, there is increasing evidence that a significant fraction of atmospheric soot is neither compact nor uniformly coated. We have generated soot particles with different initial mixing states and investigated morphological changes occurring during aging for different coating materials and aging environments. The mixing state and morphology of soot aggregates were determined from a combination of mass-mobility measurements and environmental electron microscopy imaging. The results obtained in this study will be discussed in connection with current pressing needs to identify and evaluate key parameters that control the mixing state and structure of soot nanoparticles during aging, determine the mechanism and timescale of morphology restructuring, and evaluate associated changes in cloud-forming and optical properties of particles. A better understanding of soot aging will help to reduce uncertainties in climate prediction because soot from incomplete combustion of fossil fuels is a significant contributor to direct climate forcing.

J4.4

Structure-Reactivity Analyses for Epoxide, Organosulfate, and Organonitrate Reactions Relevant to Secondary Organic Aerosol Composition

Matthew J. Elrod, Oberlin College, Oberlin, OH

Extensive studies of secondary organic aerosol (SOA) formation in the southeastern United States have identified epoxide intermediates as key species in the formation of isoprene-derived SOA. In this mechanism, SOA species are formed from the isomerization and nucleophilic addition reactions of common SOA nucleophiles (such as water and sulfate) with epoxides formed from gas phase oxidation of isoprene and related species. In this study, experimental laboratory kinetics data were analyzed to ascertain the structure-reactivity relationships that rationalize the kinetics results for a variety of systems, including isoprene-derived epoxides (IEPOX) and methacrolein-derived epoxide (MAE). Epoxide substituent effects were used to rationalize the differing nucleophilic addition mechanisms and reaction rate constants, and a nucleophilicity scale was developed to rationalize product formation observations under conditions in which two or more nucleophiles are present. A similar structure-reactivity analysis was performed for organosulfate and organonitrate species that may undergo reaction under SOA conditions. These analyses may form the basis for predicting the SOA reactivity of as-of-yet unidentified epoxides, organosulfates, and organonitrates in the presence of the variety of nucleophiles known to exist on SOA.

J4.5

Isoprene Suppression of New Particle Formation: Potential Mechanisms and Atmospheric Implications

Shan-Hu Lee, University of Alabama in Huntsville, Huntsville, AL

Secondary aerosols formed from anthropogenic pollutants and natural emissions have substantial impacts on human health, air quality and the Earth's climate. New particle formation (NPF) contributes up to 70% of the global production of CCN, but the effects of biogenic VOCs and their oxidation products on NPF processes are poorly understood. Observations have shown that isoprene, the most abundant biogenic species, suppresses NPF in forests. But the previously proposed chemical mechanism underlining this suppression process contradicts atmospheric observations. Here, we provide new insights on isoprene suppression of the biogenic NPF, based on comprehensive observations of key chemical precursors in a rural forest in the Southeastern U.S. and quantum chemical calculations. Our findings imply that in an isoprene-dominant forest, volatile oxidation products formed from isoprene compete with low-volatility oxidation products from monoterpenes in clustering with sulfuric acid, to suppress the growth of clusters. Current climate models treat NPF processes by considering only sulfuric acid and total low-volatility organic compounds, regardless of forest biogenic emission patterns over the globe. We conclude that NPF suppression in isoprene-emitting forests should be included in models to correctly predict the climate forcing by aerosols and clouds.

J4.6

Growth of Freshly Nucleated Particles by Condensation of Low-Volatility Organics

Neil M. Donahue, Center for Atmospheric Particle Studies, Carnegie Mellon University, Pittsburgh, PA; and W. Chuang, J. Troestl, and D. Worsnop

New-particle formation involving organic compounds may play an important role in climate. Here we describe measurements at the CERN CLOUD chamber facility in which new-particle formation and growth was driven by oxidation products of alpha-pinene in the absence of inorganic species. We were able to model the growth rate of these freshly nucleated particles between 1 and 30 nm using observed distributions of highly-oxidized low-volatility organic compounds within the framework of the Volatility Basis Set. We shall discuss implications for current and pre-industrial particle formation and climate.

J7.1

A Multi Isotope Approach to Understanding Aerosol Formation, Transformation and Climate: From 3.8 Billion Years Ago to Present. Earth and Mars

Mark Thiemens, University California San Diego, La Jolla, CA

The formation of atmospheric aerosols, their chemical transformation and perturbation on the earth's climate and oxidation state have been recognized for decades. Chemistry on the liquid layer of the aerosols is also an important process but notroioulsy difficult to measure. An array of experimental and theoretical approaches on achieving a maximal understanding of their properties, chemical changes, and interaction with the global and local environment is mandatory, especially to predict future impact and potential consequences of the earths atmosphere-surface environment.

The use of isotope ratios of aerosols have been done for decades and new information gathered, particularly for sulfate. In the case of sulfate, with 4 stable isotopes of sulfur, three isotopes of oxygen, and a naturally produced 35S isotope (87 day half life) an enormous amount of chemical oxidative information is stored, including ozone/oxygen chemistry in the troposphere and stratosphere is available. We have developed all techniques to measure all of these systems on a single sample. The sulfur isotopes yield information on both the source and photochemistry of its precursor sulfur dioxide, especially to the stratosphere, such as El Nino, volcanoes, and massive biomass burning processes. The inclusion of measurements of the 35S radioactivity in SO2 and SO4, a chronometer of the actual gas to particle process is available to determine the actual rate. This is the only clock available to directly measure the nucleation rate of sulfate aerosols, or any aerosol. The half life of 87 days is exactly right to make these measurements and is extremely sensitive.

Using this array of isotope measurements, coupled with basic laboratory measurements of the fundamental physical chemical processes, deeper understanding of a variety of process are studied. This includes long range transport (Asia-California; equator to poles), oxidation/nucleation rates, stratospheric chemical perturbations from volcanoes over the past 400 years, and definition of the interaction of aerosols on the overall oxidation capacity of the Earth. Measurements of samples from the Tibetan plateau have shown how isotopes probe deeper into details of the stratosphere-troposphere exchange processes, and the impact of aerosols on the rate of ice melting in the glaciers supplying 65% of the worlds drinking water. The same measurement in aerosols that later are incorporated into the ice and rock record are stored and offer views into the recent (decadal) and past (3.8 billion year)and provide climate and atmospheric information that could not have been acquired by any other technique. Ice core measurements across the Holocene detail the oxidation state changes through glaciation periods. The impact on climate by volcanoes (such as the year of no summer) have also been detected as well as the details of the chemical perturbation of Pinatubo, El Chichon and Cerro Hudson. Such information are critical in providing the deepest understanding of the Earth's upper atmosphere

This presentation will describe the use of the wide isotopic measurements of multi isotopes in determining global atmospheric chemistry, interaction with aerosols, and our development of understanding the perturbation history of the Earth's climate. The understanding of the Earths system is also understood by comparing to the same formation and transformation process in another natural environment, Mars, and its change over the same time scale.

J7.2

Chemical Reactivity and Non-Liquid/Liquid States of Secondary Organic Material

Scot T. Martin, Harvard University, Cambridge, MA; and Y. J. Li, A. K. Bertram, P. Liu, Z. Gong, Y. Wang, A. P. Bateman, and C. Bergoend

The reactivity of secondary organic material (SOM) of variable viscosity, ranging from non-liquid to liquid physical states, was studied. The SOM was produced as particles of 50 to 150 nm in diameter in aerosol form from six precursors, including three terpenoid and three aromatic species. The viscosity of the hygroscopic SOM was adjusted by exposure to relative humidity (RH) from <5% to >90% RH in steps of 10% at 293 +/- 2 K. The aerosol was subsequently exposed to 5 ppm NH3 for mean reaction times of 30, 370, or 5230 s. The ammonium-to-organic ratio of mass concentrations in the particles increased monotonically from <5% RH to a limiting value at a threshold RH, implicating a switchover in the reaction kinetics from a system limited by diffusivity within the SOM for low RH to one limited by other factors, such as saturated uptake, at higher RH. The switchover RH for reactivity differed in all cases from the transition RH of a non-liquid to a liquid state.

The substantial differences in RH for transitions in chemical reactivity compared to those of physical state imply that chemical transport models should not use the non-liquid to liquid transition of SOM as a one-to-one surrogate for SOM reactivity.

J7.3

Contributions of Amines and Highly Oxidized Organics to New Particle Formation in the Atmosphere: Global Simulations and Implications

Fangqun Yu, State University of New York, Albany, NY; and G. Luo

Particle number concentration is an important factor in the health and climate impacts of atmospheric aerosols. Secondary particles formed via new particle formation (NPF) or nucleation dominate the global total particle number abundance, contributing as high as 80-95% to total particle (larger than ~3 nm) and 50-80% to cloud condensation nuclei (CCN) number concentrations in most parts of the troposphere. Recent laboratory measurements indicate important roles for amines and highly oxidized low volatility secondary organic gases (LVSOG) in NPF in experimental chambers but the actual role of these species in NPF in the real atmospheric remains unclear. Here, we investigate the possible contributions of amines and LVSOG to atmospheric NPF, using a state-of-the-art global chemical transport model (GEOS-Chem) with an advanced particle microphysics (APM) model incorporated. Schemes to calculate the concentrations of amines and LVSOG as well as nucleation rates involving these species have been implemented into GEOS-Chem/APM. For particle growth, the model considers kinetic condensation of H2SO4 and LVSOG, equilibrium uptake of nitrate and ammonium, partitioning uptake of relatively high volatility secondary organic gases, and particle self-coagulation. The particle number concentrations predicted based on LVSOG-H2SO4 and amines-H2SO4-H2O nucleation parameterizations are compared to those based on the binary ion-mediated nucleation (IMN) of H2SO4-H2O as well as to relevant field measurements. We show that both LVSOG-H2SO4 and IMN schemes capture the observed high frequency of NPF in spring, but the LVSOG-H2SO4 scheme significantly over-predicts while the IMN scheme slightly under-predicts NPF and particle number concentrations in summer. Amines-H2SO4-H2O nucleation is significant in the boundary layer in Asia, central Europe, and some parts of America but is small above boundary layer and over oceans. Key parameters controlling nucleation rates based on different schemes, uncertainties, implications, and future research needs will be discussed.

J7.4

Henry`s Law Constant of Atmospheric Amines and their Dependence on Temperature, pH, and Ionic Strength

Yong Liu, University of Colorado Dener, Denver, CO; and C. Leng and J. Roberts

In the atmosphere, amines are emitted from various anthropogenic (automobiles, industries, combustion, animal husbandry, cooking, tobacco smoke, treatment of sewage and waste, and CO2 capture), biogenic (ocean, biomass burning, and vegetation), and geologic sources. Atmospheric amines are basic, so they have acid-neutralizing capacity and potential to engage in acid-base reactions in the condensed phase, which could contribute to formation of new aerosol particles. Atmospheric amines can also participate in cloud chemistry and contribute to brown carbon formation. Despite the importance, amines are usually not incorporated into regional and global atmospheric chemical transport models largely due to lack of their thermodynamic properties such as The Henry`s law constants (KH). In this work, we choose six different amines and measure their KH in pure water and in 1-octanol for the temperatures pertinent to the lower troposphere (278 - 298 K) using a bubble column system coupled to an FTIR spectrometer. The KH values display strong dependence on temperature, pH and ionic strength. The characteristic time for amines to establish gas-droplet equilibrium is relatively short, and it can be significantly extended at lower temperatures or pHs. The evaluation of amines partitioning between gas phase and condensed phase implies that amines predominantly resides in rainwater, and loss to organic aerosol is negligible.

J7.5

Size-Resolved Measurements of Ice Nucleating Particles at Six Locations in North American and One in Europe

Ryan H. Mason, University of British Columbia, Vancouver, BC, Canada; and M. Si, C. Chou, V. Irish, R. Dickie, P. Elizondo, R. Wong, M. Brintnell, M. Elsasser, W. Lassar, K. Pierce, K. Suski, J. P. D. Abbatt, P. J. DeMott, T. C. J. Hill, J. A. Huffman, W. R. Leaitch, A. M. Macdonald, A. Platt, R. Sarda-Estève, C. L. Schiller, D. Toom-Sauntry, and A. K. Bertram

Ice nucleating particles (INPs) are a small fraction of the total aerosol population capable of catalyzing ice formation under atmospheric conditions. The formation, albedo, and lifetime of mixed-phase and cirrus clouds may be influenced by the presence of such particles. Compared to ambient measurements of the total number concentration of INPs, relatively little data exists on the size distribution of INPs in the atmosphere. This size information may be useful in source identification, modeling the transport of INPs in the atmosphere, and for determining if common instrumentation captures the majority of the atmospheric INP population. Measured using the micro-orifice uniform deposit impactor-droplet freezing technique (MOUDI-DFT), we report immersion-mode INP number concentrations as a function of particle size at several ground-level sites across North America and Europe, including Arctic, alpine, coastal, marine, agricultural, and suburban environments. More than 91 % of INPs active at -15 °C were found to be supermicron in size and 62 % were in the coarse mode (> 2.5 μm). While these percentages decreased with decreasing temperature, supermicron INPs remained important with nearly half of those active at -25 °C belonging to the coarse mode.

J8.1

Meeting the Climate Challenge of 2 Degrees Centigrade: A Study Using an Integrated Economics and Earth System Model

Ronald G. Prinn, MIT, Cambridge, MA; and A. Sokolov and S. Paltsev

The global average temperature has increased by almost 0.9 degC between 1880 and 2014. The evidence is now very compelling that most of this increase is due to human activity, especially activity that produces carbon dioxide and many other greenhouse gases (GHGs). There are now also compelling reasons, summarized by the IPCC 5th Assessment (2013), to regard a warming of about 2 degC between 1880 and 2100 as a threshold, above which the damages globally to human and natural systems due to climate change begin to become economically and ethically less and less tenable and damages above 3 degC warming are certainly untenable. But there is also an equally compelling case for sustaining economic growth in developed and developing nations. Also, the uncertainties in climate projections mean that we must think in terms of the probability, not certainty, of reaching these thresholds. We present results using the MIT Integrated Global System Model (http://globalchange.mit.edu) exploring combinations of low and zero-emission technologies and their costs over time that achieve the 2 and 3 degC targets with at least 50% probability. The IGSM is well suited for this study. The IGSM has two coupled sub-models: the MIT Economic Projection and Policy Analysis (EPPA) model and the MIT Earth System Model, that simulate human activity and Earth System response to this activity respectively. The EPPA simulates the evolution of the major economic, demographic, trade, and technological processes involved in the production of GHG and aerosol emissions relevant to climate and air pollution at national and global levels. It has detailed considerations of all relevant energy, industrial, agricultural and transportation sectors and uses comprehensive regional data for production, consumption and trade. The EPPA emissions of all major GHGs covered by the Kyoto and Montreal Protocols, and primary and secondary air pollutant emissions that lead to radiatively active aerosols and ozone, are inputs into the MESM, an earth system model of intermediate complexity, which has coupled sub-models of atmospheric and oceanic dynamics, atmospheric chemistry, terrestrial and oceanic biogeochemistry, terrestrial bio-geophysics, and physics of the ocean and land cryosphere. For our study, detailed chemistry and radiative forcing calculations for each major GHG and aerosol are included in MESM. To address important climate and air pollution interactions, emissions in urban areas are first input into an urban-scale air chemistry module within MESM, while those outside urban areas are input into the global MESM model. We find that as the price imposed on emissions rises, the fraction of energy from low and zero emission technologies rises relative to fossil. Also as the carbon price increases, the energy use decreases due to higher energy efficiency. The current uncertainty in the response or sensitivity of the climate system to carbon dioxide and other greenhouse gases strongly affects the needed emission prices and the resultant scales of the changes in primary energy sources and efficiency in energy use over time. For a high climate response, the average global energy use is about 30% less, the fraction of energy production from renewables is about 40% more, and the required emissions price is about 3 times higher, compared to a low climate response. The competition among these low emission technologies is largely determined by their assumed relative costs, while the total cost is determined largely by the optimism about future technologies and policy instruments chosen to achieve the 2 or 3 degC target.

J8.2

Projection of Future Air Quality in South Asia Using High Resolution

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About one seventh of the world's population living in South Asia faces the risk of severe air pollution as anthropogenic emissions of air pollutants are increasing continuously due to the economic development and urbanization in the region. Recent studies show that air pollution in South Asia is already reducing the lifespan of about 660 million people by about 3 years and destroying enough food to feed about 94 million people. This problem may worsen in the future as anthropogenic emissions are expected to increase further due to economic growth in South Asia, and climate change is expected to lead to atmospheric conditions conducive for the production and accumulation of air pollutants. To learn how air quality will change in South Asia from present day conditions to the 2050 future time frame, we are conducting high resolution air quality simulations for the present day (1995-2005) and future (2046-2055) time periods over South Asia using the Nested Regional Climate Model coupled with Chemistry (NRCM-Chem). The model domain covers entire South Asia at a resolution of 60 km with a nested domain over the densely populated and polluted Indo-Gangetic Plain region at a resolution of 12 km. The model results are being evaluated with available in situ and satellite based observations and the evaluation results show that that NRCM-Chem model is able to capture several important features of the observed spatial and temporal distribution of key meteorological parameters and air pollutants. Preliminary analysis of model results show that annual average surface ozone and PM2.5 may increase by up to 15 ppbv and 25 µg m-3, respectively under RCP8.5 scenario with highest increase in the Indo-Gangetic Plain.

J8.3

Mario Molina's Research Not Only Led to the Solution for the Antarctic Ozone Hole, But it Led to the Solution for Global Warming Caused by Humans Since 1960

Peter L. Ward, USGS Retired, Jackson, WY

The ozone layer protects life on Earth by absorbing ultraviolet-B solar radiation, the most energetic, hottest, solar radiation to regularly reach the lower stratosphere. Ultraviolet-B is harmful because, at adequate dosage, it burns human skin, causes skin cancer, and degrades materials left outdoors. Ultraviolet-B radiation causes photodissociation of ozone. When there is less ozone in the lower stratosphere than normal, more of this very hot radiation is observed to reach Earth. Chlorofluorocarbon gases (CFCs) became widely manufactured in the 1960s for use as refrigerants, spray-can propellants, and fire-retardants. By 1970, ozone depletion began to increase, especially in polar regions where it is enhanced by polar stratospheric clouds. Mean surface air temperatures also began to increase, especially in the northern hemisphere, where human-enhanced, ground-level ozone concentrations were heated by photodissociation caused by increasing ultraviolet-B radiation penetrating to Earth's surface. As a result of the Montreal Protocol, emissions of CFCs stopped increasing in 1993, ozone depletion stopped increasing in 1995, and mean surface air temperatures stopped increasing in 1998. Ultraviolet heat absorbed on land is primarily radiated back into the atmosphere at night as infrared radiation, but ultraviolet radiation penetrates oceans to depths of tens of meters, where the heat is not radiated back into the atmosphere until the whole ocean surface warms. Oceans store 1000 times more heat than the atmosphere. Since ozone depletion is expected to continue for decades, ocean temperatures will continue to rise and glacial ice will continue to melt. The fundamental problem with greenhouse-gas theory is with the way thermal energy has been calculated since the 19th century. Based on equations by Maxwell (1865) and mathematics developed in the 1890s, electromagnetic radiation is thought to propagate as waves where energy is a function of wave-amplitude squared as well as the bandwidth of the wavelengths absorbed. But there is no physical way for waves to travel through space. Michelson and Morley (1887) showed that there is no medium for waves to travel in. Computer models based on greenhouse-gas theory predict that mean surface air temperatures should continue to increase after 1998 because concentrations of carbon dioxide have continued to increase. These computer models also calculate that Earth is warmed more by infrared radiation from Earth than by ultraviolet radiation from Sun. Yet it is common knowledge that one gets much hotter standing in sunlight bathed in ultraviolet radiation, than standing outdoors at night, bathed in infrared radiation. Small amounts of ultraviolet radiation burn skin, while endless exposure to infrared radiation cannot burn skin and cannot even energize photosynthesis. The second law of thermodynamics states, in effect, that thermal energy can only flow from higher temperature to lower temperature. No heat can flow spontaneously by conduction or by radiation from a colder body to a warmer body. Layers of gas in the atmosphere are observed to be colder than Earth's surface. Even if these gases do radiate infrared energy back towards Earth, this colder radiation is reflected, not absorbed, by a warmer Earth. One does not stand next to a cold stove to get warm. When you heat matter, the frequencies and amplitudes of very high-frequency oscillations of all the degrees of freedom of all the bonds that hold matter together are observed to increase. Planck's law shows that with increasing temperature of matter, the amplitudes of these oscillations increase at every frequency and the frequency of the highest amplitude oscillations also increases. Heat in matter is stored in these oscillations since the heat capacity of matter increases with increasing number of degrees of freedom of the bonds. Heat flows in matter from higher amplitude of oscillation to lower amplitude of oscillation via resonance. Heat flows into a colder atmosphere or space when these oscillations on the surface of matter induce electromagnetic oscillations that radiate the heat through air or space. This radiation, when absorbed by colder matter, increases the amplitudes of bond oscillations via resonance, warming the matter. The thermal energy of these oscillations at each frequency is equal to the frequency times the Planck constant. Thermal energy of ultraviolet-B radiation reaching Earth when ozone is depleted, is 48 times greater, 48 times hotter, than infrared radiation absorbed most strongly by carbon dioxide. The air temperature at Earth's surface is determined primarily by the dosage of thermal energy reaching Earth. Changes in surface temperature are likewise caused by changes in the dosage of thermal energy reaching Earth. The amount of ultraviolet-B radiation not absorbed by the ozone layer, has the greatest effect on changing Earth's surface temperature. The effect of infrared radiation absorbed by greenhouse gases is, at least, 48 times smaller. Ozone is also depleted by chlorine and bromine emitted from active volcanoes of all sizes. Effusive, basaltic volcanic eruptions, typical in Hawaii and Iceland, extrude large lava flows, depleting ozone and causing global warming. Major explosive volcanoes, on the other hand, also deplete ozone but in addition eject megatons of water and sulfur dioxide into the lower stratosphere where they form sulfuric-acid aerosols whose particles grow large enough to reflect and scatter ultraviolet sunlight, causing net global cooling for a few years. The relative amounts of explosive and effusive volcanism are determined by the configuration of tectonic plates moving around Earth's surface. Detailed studies of climate change throughout geologic time are not well explained by greenhouse-gas theory, but are explained quite clearly by ozone-depletion theory. Ozone concentrations vary substantially by the minute and show close relationships to synoptic highs and lows, as pointed out by Dobson in the 1920s, the height of the tropopause, and to the strength and location of polar vortices and jet streams. Integrating the effects of volcanism on ozone concentrations and the effects of ozone concentrations on synoptic weather patterns should improve weather forecasting. For example, the volcano Bárgarbunga, in central Iceland, extruded 85 km2 of basaltic lava between August 29, 2014, and February 28, 2015, having a profound effect on weather. Details are explained in my new book "What Really Causes Global Warming? Greenhouse gases or ozone depletion" and at ozonedepletiontheory.info.

J8.4

Climate Change: Science, Policy and Solutions

Mario J. Molina, University of California, San Diego, San Diego, CA

Climate change is the most serious environmental challenge facing society in the 21st century. The basic science is clear: the average temperature of the Earth's surface has increased about 0.8 degrees Celsius since the Industrial Revolution, and the frequency of extreme weather events such as heat waves, droughts and floods is also increasing, most likely as a consequence of this temperature change. There are scientific uncertainties that remain to be worked out, connected with issues such as the feedback effects of clouds and aerosols. Nevertheless, the consensus among experts is that the risk of causing dangerous changes to the climate system increases rapidly if the average temperature rises more than two or two and a half degrees Celsius, and that the associated costs to the economy are likely to be significantly larger than the costs associated with adopting the necessary measures to minimize such risk. The scientific community needs to better communicate the nature of the climate change risk to decision makers in government to improve the likelihood of reaching an international agreement that effectively reduces global greenhouse gas emissions.

J9.1

Measuring Sources and Sinks of Greenhouse Gases at The "Missing Scales: Ecosystem to Continent, Weeks to Decades

Steven Wofsy, Harvard University, Cambridge, MA

Increases in the concentrations of methane (CH4) and carbon dioxide (CO2) are powerful drivers of global change and climate warming. In order to understand the underlying causes and to design mitigation strategies, the emissions of these gases must be quantified, and the response to the driving biological, physical, and human processes must be understood. The key spatial scales for this analysis range from ecosystems, landscapes, urban, regional, to continental, and the temporal scales range from seasonal to decadal. Direct observations of sources and sinks are very challenging on these scales. In this talk, case studies focusing on emission of CH4, in the Arctic and the Continental US, will be presented illustrating why these spatial and temporal scales are so important, and showing how new approaches that combine direct observations with model-data assimilation methods provide the means to determine greenhouse gas fluxes at the "missing" scales with growing confidence and accuracy.

J9.2

Observing the Anthropocene from Instrumentation on Aircraft and Space: (from SCIAMACHY to CarbonSat and SCIA-ISS)

John P. Burrows, University of Bremen, Bremen, Germany

From the Neolithic revolution to the industrial revolution over ~ 10 000 years, the earth's population rose from several millions to 1 Billion powered by energy from a mixture of biofuels, water and solar power and some coal burning. Following the industrial revolution, which began in the UK in the late 18th century, fueled by the combustion of fossil fuels, a dramatic rise in both the human population, now comprising over 7 Billion with more than 50% living in urban areas, and its standard of living has occurred. The expectation is that by 2050 population will be of the order of 10 Billion with 75% dwelling in urban areas. Anthropogenic activity has resulted in pollution from the local to the global scale, changes in land use, the destruction of stratospheric ozone, the modification of biogeochemical cycling, the destruction of species, ecosystems and ecosystem services and climate change. The observation of atmospheric composition provides a unique early warning of the natural and anthropogenic origins of change. Consistent and consolidated measurements from the local to the global scale are required to test our knowledge of the biogeochemical cycle determining atmospheric composition and dynamics and to assess and attribute accurately their modification by anthropogenic activity. To achieve global measurements of atmospheric constituents (trace gases, aerosol and cloud parameters) the SCIAMACHY (SCanning Imaging Absorption spectroMeter for Atmospheric CHartographY), was initiated in the early 1980s. This was the first passive remote sensing space based instrumentation, designed to make simultaneous contiguous measurements of the solar upwelling radiation at the top of the atmosphere from the ultraviolet visible to the shortwave infrared. The SCIAMACHY project resulted in measurements of the instruments GOME, originally called SCIA-mini, on ESA ERS-2 (1995 to 2011), SCIAMACHY on ESA Envisat (2002 to 2012), GOME-2 on ESA/EUMETSAT Metop series (2006 to 2020) as well as the planned EU Copernicus/ ESA /EUMETSAT Sentinel 4, originally called GeoSCIA, which will be the first geostationary instrument of its kind flying on Meteosat Third Generation Sounder from 2019 to 2015 to 2034 and the Sentinel 5, which is the follow on to GOME-2 and will fly on Metop Second Generation from 2020 to 2035. The GOME/SCIAMACHY/GOME-2 observations have been successfully used to retrieve information about trace tropospheric constituents. These include the column amounts of O3, NO2, HCHO, CHO.CHO, CO, H2O, IO, BrO, the dry mole fraction of CO2 and CH4, as well as aerosol and clouds parameters, ocean colour and sun induced fluorescence from space. This presentation will address key issues related to our understanding of the changes of the above parameters. Similarly the evolution of the observing system through the development the demonstration of aircraft precursors instruments, such as AIRDOAS and MaMap for CarbonSat and SCIA-ISS.

J9.3

Measurements of Bromine Monoxide and Iodine Monoxide in the Lower Stratosphere: Constraints on Total Inorganic Halogen Budgets and Chlorine Activation

Theodore Koenig, University of Colorado, Boulder, CO; and R. Volkamer, S. Baidar, B. Dix, E. Atlas, S. Schauffler, V. Donets, R. Lueb, M. Navarro, E. C. Apel, N. J. Blake, A. Hill, R. Hornbrook, D. D. Riemer, A. J. Weinheimer, S. Hall, K. Ullmann, T. Hanisco, G. M. Wolfe, D. Anderson, T. L. Campos, M. Reeves, P. Romashkin, L. L. Pan, R. J. Salawitch, S. Y. Wang, A. Saiz-Lopez, R. P. Fernandez, D. Kinnison, J. F. Lamarque, D. J. Jacob, J. Schmidt, M. Evans, and T. Sherwen

Halogens have important direct and indirect effects on atmospheric chemistry e.g. ozone destruction, oxidative balance, mercury processing, particle seeding and growth. The monoxides of bromine and iodine (BrO and IO) are important constituents of inorganic halogens and additionally can activate chlorine through interhalogen reactions. These species were measured by the Airborne Multi AXis Differential Optical Absorption Spectroscopy (AMAX-DOAS) instrument during the CONvective TRansport of Active Species in the Tropics (CONTRAST) campaign. Here we examine two case studies – CONTRAST RF06 and RF15 – which sampled the tropical UTLS during a horizontal transect jet-crossing into the mid-latitude lower stratosphere. Our measurements include the first detection of IO in the lower stratosphere. We use a chemical box-model – constrained by measurements of BrO and IO as well as by AWAS, TOGA, and other measurements on the aircraft – to determine the total budgets of inorganic bromine and iodine. We examine the implications for ozone destruction both directly and through activation of chlorine, and we further investigate correlations of total Bry and Iy with air mass indicators such as CFC-11.

J9.4

Tropical Tropospheric Bromine and Stratospheric Injection from VSL Compounds Inferred from CONTRAST and ATTREX

Ross J. Salawitch, University of Maryland, College Park, College Park, MD; and P. Wales, J. M. Nicely, D. C. Anderson, T. P. Canty, S. Choi, J. P. Joiner, E. Atlas, S. Schauffler, V. Donets, R. A. Lueb, M. Navarro, E. C. Apel, N. J. Blake, A. Hills, R. Hornbrook, D. D. Riemer, D. Chen, L. G. Huey, D. Tanner, R. Volkamer, T. Koenig, S. Baidar, B. Dix, A. J. Weinheimer, G. M. Wolfe, T. Hanisco, S. Hall, K. Ullmann, R. P. Fernandez, A. Saiz-Lopez, D. E. Kinnison, J. F. Lamarque, S. Honomichl, and L. L. Pan

The CONvective TRansport of Active Species in the Tropics (CONTRAST) and Airborne Tropical Tropopause Experiment (ATTREX) field campaigns were designed in part to quantify the abundance of very short lived (VSL) bromocarbons in the marine boundary layer (MBL) of the Tropical Western Pacific, the variation with respect to altitude of these compounds from the MBL to the base of the tropopause transition layer, and the abundance of BrO throughout the tropical troposphere. Here we examine the theoretical understanding of VSL source gases by comparing measurements of these bromocarbons provided by two instruments, AWAS and TOGA, to model values found using CAM-Chem. We examine also the bromine budget in the tropical troposphere: i.e., consistency between tropospheric loss of these compounds and the appearance of products using observations of BrO from two other instruments, CIMS and DOAS. Finally, implications for supply of bromine to the lower stratosphere via source gas and product gas injection will be examined, based largely on data collected during flights that probed the extra-tropical, lower stratosphere.

J9.5

Direct Radiative Forcing of Black Carbon Particles: A Global Climate Model Perspective

Yuan Wang, California Institute of Technology/JPL, Pasadena, CA; and J. Peng, M. Hu, and R. Zhang

Black carbon (BC) particles in the atmosphere have profound impacts on air quality, human health, weather, and climate. Although BC represents a key short-lived climate forcer, its direct radiative forcing remains highly uncertain. In particular, the results of absorption enhancement of BC particles during atmospheric aging from available in-situ measurements and laboratory studies are conflicting, leading to large uncertainty in global radiation budget and climate variability. Using a global climate model, the BC forcing change due to the mass absorption cross-section (MAC) enhancement is evaluated, on the basis of the most recently available experimental and field studies. Moreover, the observed MAC enhancement associated with accumulation mode aerosols is implemented in NCAR CAM5. The forcing calculations include both direct radiative forcing and forcing due to the cloud adjustment, in particular the BC semi-direct effects. Atmospheric implications of our results are discussed.

J9.6

Combining Laboratory Studies and Radiative Transfer Modeling to Better Assess the Risks of Geoengineering Using Stratospheric Aerosols

John A. Dykema, Harvard University, Cambridge, MA; and F. N. Keutsch and D. W. Keith

Discussions of technological solutions that could complement essential emissions cuts to mitigate climate change in the coming decades have become significantly more prominent in the scientific discourse in recent years. One proposal that has received significant attention is to utilize sulfate aerosols in the stratosphere to scatter solar radiation back to space. It is known that this proposal carries multiple risks, including ozone loss, dynamical changes to the stratosphere, and changes in the ratio of diffuse to direct solar radiation. Other types of particles might plausibly reduce these negative impacts relative to sulfate aerosols. For example, there are studies analyzing the use of solid aerosol particles, such as alumina and titania. Furthermore, the optical properties of other materials may feature a reduced infrared heating rate, and a more favorable ratio of backwards scattering to forward scattering. Because volcanic eruptions can inject sulfate aerosols directly into the stratosphere, there is a substantial body of scientific knowledge to analyze how sulfate aerosol injection would affect the stratosphere. Any consideration of novel materials, such as solid aerosol particles, would require a rigorous assessment of chemical and optical properties as a starting point for further discussions.

Laboratory experiments would play a primary role in such assessments. Studies to quantify the rates of heterogeneous reactions, such as the activation of halogens, on solid aerosols require careful design to control the interaction of gas flows with chamber walls and to measure reaction products with adequate sensitivity. Ideally, these experiments would consider the impact of UV illumination to analyze photocatalytic effects on gas- and liquid-phase constituents when illuminated by a realistic spectral distribution relative to the sun. We will present a new flow tube system under development in our research group and radiative transfer modeling results that, when combined, will provide a quantitative framework for assessing the risks and efficacy of solid particles for geoengineering applications.

J10.1

Mario J. Molina: Creative Science that Redefined Human Responsibility to the Planet While Uniting Chemistry and Meteorology

James G. Anderson, Harvard University, Cambridge, MA

Mario Molina's research, extending over more than four decades, stands as a triumph of human thought bridging from a molecular level understanding of kinetics, photochemistry and catalysis to a series of globally critical insights central to our understanding of stratospheric ozone that underscored the delicate nature of life at the planet's surface. As a result of the now recognized sensitivity of human health to the column concentration of stratospheric ozone, Mario's initial work on chlorofluorocarbons (CFCs) with Sherry Rowland redefined an understanding of global toxicology. But his insight into chemical mechanisms that lead to the Molina chlorine dimer mechanism provided the key foundation for dissecting the cause of the Antarctic Ozone Hole. This led directly to the formulation and international acceptance of the Montreal Protocol banning the production of CFCs. It was subsequently demonstrated that the chlorine dimer mechanism is responsible for the dramatic loss of ozone over the Arctic in late winter and early spring. However, Mario's contributions now link catalytic chemistry to meteorology through a combination of factors that place the summertime lower stratosphere over the U.S. in a unique position at the intersection of climate forcing and potential ozone loss in the stratosphere. The factors that conspire to establish this circumstance engage four independent considerations. First is the occurrence of severe storms in the U.S. Mid-West caused by the intersection of moist airflow from the Gulf of Mexico with the subtropical jet in late spring and summer. These storms are characterized by a combination of tornadoes, hail, heavy precipitation and high winds, the frequency and intensity of which are increasing with increased forcing of the climate system by the addition of CO2, CH4, N2O CFCs, and other infrared active species to the atmosphere associated with human activity. Second is the recognition that these severe storms are capable of injecting water vapor deep into the stratosphere over the U.S., with injection depths reaching the altitude of increasing inorganic halogen species formed by the photolytic breakdown of organic chorine and bromine transported into the stratosphere. Third is the recognition that the catalytic conversion of inorganic halogen species to chlorine and bromine radicals occurs on ubiquitous sulfate-water aerosols wherever and whenever the temperature-water vapor conditions are met. These conditions are met in the summertime lower stratosphere over the U.S. by virtue of temperatures between 200 and 205K in combination with convectively injected water vapor concentrations that reach 15 ppmv or greater. Fourth, is the recognition that the flow pattern of the lower stratosphere over the central U.S. in summer is repeatedly under the control of the North American monsoon that forms an anti-cyclonic (clockwise) gyre that captures the combination of convectively injected water vapor and boundary layer radical precursors within it, retaining that chemical mix for periods up to three weeks during the months of July and August. It is within this gyre over the U.S. that the catalytic reactions can occur potentially reducing the column concentration of ozone in summer as they do over the Arctic in winter. Recent developments linking observations from in situ aircraft, satellite and NEXRAD weather radar are reviewed and placed in the context of Mario Molina's seminal contributions linking the physical sciences to societal objectives.

J10.2

A Tropical Tropospheric Source of High Ozone/Low Water Filaments

Daniel C. Anderson, University of Maryland, College Park, MD; and J. M. Nicely, R. J. Salawitch, T. P. Canty, R. R. Dickerson, T. Hanisco, G. M. Wolfe, E. C. Apel, E. Atlas, T. Bannan, S. Bauguitte, N. J. Blake, J. Bresch, T. L. Campos, L. Carpenter, M. Cohen, M. Evans, R. P. Fernandez, B. H. Kahn, D. E. Kinnison, S. Hall, N. R. P. Harris, R. Hornbrook, J. F. Lamarque, M. Le Breton, J. Lee, C. Percival, L. Pfister, R. B. Pierce, D. D. Riemer, A. Saiz-Lopez, A. M. Thompson, K. Ullmann, A. Vaughn, and A. J. Weinheimer

Numerous field campaigns have noted the prevalence of filaments of high O_3 and low H_2O (HOLW) in the tropical western Pacific (TWP) troposphere. These filaments can drastically alter the meteorology, climate, and chemistry of the region through suppression of convection, changes in local radiative forcing, and alteration of the OH budget and are often cited as having a dynamical origin caused by transport from either the mid-latitude upper troposphere (mlUT) or stratosphere. We will show that HOLW filaments were a dominant feature observed during the CONTRAST and CAST field campaigns conducted in the TWP during January and February 2014. Our back trajectory analysis connects these filaments to regions of active biomass burning in the tropics, namely Africa and Southeast Asia. The low relative humidity observed in these filaments is consistent with large-scale descent in the tropics, during the transit from the source region to the TWP. The chemical composition of the filaments confirms a biomass burning origin and demonstrates negligible influence from the mlUT and stratosphere. Since the high O_3 in these filaments is of tropical origin, biomass burning likely exerts a stronger influence on earth's climate than commonly appreciated.

J10.3

Laboratory Studies to Investigate the Role of Volatile and Semi-Volatile Organic Compounds in the Atmospheric Aging of Biomass Devolatilization Products

Claire Fortenberry, Washington University in St. Louis, St Louis, MO; and M. Walker, Y. Zhang, D. Mitroo, C. Oxford, W. Brune, and B. Williams

Although biomass burning organic aerosol (BBOA) contributes a significant fraction of atmospheric organic aerosol (OA), the temporal variation in chemical composition due to oxidation has been difficult to characterize. Volatile, intermediately-volatile, and semi-volatile organic compounds (VOCs, IVOCs, and SVOCs, respectively) are co-emitted with primary OA (POA) during biomass combustion and participate in the formation of secondary organic aerosol (SOA) as biomass burning plumes age in the atmosphere. However, the chemical reaction pathways and products associated with S/I/VOC oxidation and subsequent SOA formation remain poorly understood. In this study, the role of S/I/VOCs in the atmospheric photochemical aging of biomass emissions was investigated using a Potential Aerosol Mass (PAM) flow reactor

in conjunction with a Thermal desorption Aerosol Gas chromatograph (TAG), a Proton Transfer Reaction Mass Spectrometer (PTR-MS), and a high resolution time-of-flight Aerosol Mass Spectrometer (AMS).

Primary emission products were created by devolatilizing oak wood and oak leaf samples in a custom-built emissions chamber. The biomass samples were resistively heated in the chamber to 200°C, the consistent heat pulse providing emissions that are more reproducible at the molecular level as compared with direct biomass combustion techniques. The primary material was subsequently oxidized in the PAM reactor to simulate different levels of atmospheric aging. PAM oxidation experiments were conducted on primary particles alone, with the vapor fraction of the emission product removed with a denuder prior to entering the PAM, and separately on the gas-phase material following the removal of particles with a Teflon filter. The TAG provided chemical speciation data for the oxidized aerosol, allowing for molecular level identification at each level of oxidation. Additionally, the TAG measured the thermal decomposition fragments of the most thermally labile aerosol components. The AMS was used to obtain high-resolution mass spectra and oxidation state information for the bulk aerosol, and the PTR-MS was used to measure concentrations of different gas-phase species. A comparison of the chemical properties of filtered, denuded, and unfiltered/undenuded biomass emission oxidation products will be presented in order to demonstrate the impact of S/I/VOCs on the chemical evolution of BBOA in the atmosphere. Specifically, the oxidative evolution of key molecular marker compounds will be shown in order to elucidate the chemical reaction pathways associated with SOA formation and aging of BBOA.

J10.4

Comparison of Global Tropospheric Ozone Precursors from Measurements and the MACCity Global Emissions Inventory

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Global chemistry-climate models generally have problems faithfully reproducing tropospheric ozone concentrations, seasonal cycles and interannual trends. Accurate knowledge of tropospheric ozone is important for understanding its current and future effects on human health, air quality, and climate. Successful tropospheric ozone simulations require high quality information on the emissions of ozone precursors, including nitrogen oxides (NOx), carbon monoxide (CO) and volatile organic compounds (VOCs).

We analyzed CO and NOx measurements from four megacities (Los Angeles, New York City, London, and Paris). It has been shown that CO and VOC emissions are highly correlated in urban areas. However, CO is more extensively measured than VOCs. Therefore, we compared the long-term evolution of the measured NOx/CO ratio in each city to the ratio of the emissions of these two pollutants reported by the MACCity global emissions inventory at the inventory grid points nearest the city. The longest available measurement record (~50 years) is from Los Angeles, where the measured NOx/CO ratios are consistently smaller than the emission ratio in the MACCity inventory and the slope of the long-term trend in measured NOx/CO ratios is of significantly larger magnitude than for MACCity. The other 3 cites do not have as long of a data record, but the evolution of their NOx/CO ratios also implies that the MACCity NOx/CO emissions trends are not steep enough. However, the agreement between the measured and inventory ratios is better for the short time period where measurements are available in these three cities.

Comparisons of MACCity with fuel-based emission estimates for Los Angeles (for the 1990s and 2000s) suggest that the CO emissions are underestimated in the inventory, which results in the higher NOx/CO ratios.

J10.5

Pressure-Dependent Stabilization of Carbonyl Oxides from Alkene Ozonolysis

Neil M. Donahue, Center for Atmospheric Particle Studies, Carnegie Mellon University, Pittsburgh, PA; and J. Hakala and V. Hofbauer

Carbonyl oxides (Criegee Intermediates) have gained renewed interest since an efficient source from gem-diiodoalkanes was perfected. However, the dominant atmospheric source is thought to be the stabilization of highly vibrationally excited species resulting from the extremely exothermic addition of ozone to alkenes. We have long studied the pressure dependence of this process, including prompt production of OH radicals as well production of the intermediates themselves. Here we use highly sensitive nitrate anion chemical ionization mass spectrometry to probe the pressure dependence of carbonyl oxide stabilization via the formation of sulfuric acid vapor from the reaction between carbonyl oxides and sulfur dioxide. OH radicals produced from ozonolysis also convert sulfur dioxide to sulfuric acid vapor and so we measure the ratio of sulfuric acid production with and without an OH radical scavenger to infer the ratio of OH and stabilized carbonyl oxide production. We studied pressure dependence between 30 and 900 torr of nitrogen for the reactions of ozone with a series of symmetrical trans alkenes (butene though tetradecene), 2,3 dimethyl-2-butene (tetramethylethylene) and the monoterpenes a-pinene, 3-carene, limonene, and beta-pinene.

J10.6

Analysis of TES Ozone Observations to Understand Air Pollution Transport

Ryan M. Kladar, NOAA, Boulder, CO; and O. R. Cooper

To better understand the causes of ozone formation and transport, we create and analyze global satellite ozone retrieval products for ground level to upper tropospheric ozone concentrations over the years 2005 to 2013 using the Tropospheric Emission Spectrometer (TES) that rides aboard the NASA Aura satellite. Many global and regional tropospheric ozone trends are not fully understood. Observing many different pressure levels between 1000 hPa to 215 hPa, we focus on the areas where model and other observation strategies disagree, namely the Arabian Peninsula, the Australian outback, and the southern Sahara. We observe (and these areas may be experiencing) unusually high ozone concentrations. We also comment on the historically high ozone areas such as China, Northern India, and the western and southern United States and how known phenomena compare to our observations. Many observations confirm known mechanisms of ozone formation and transport, such as the effect of the yearly monsoon cycle in South, Southeast, and East Asia. Others, such as the surprisingly high monthly average concentrations on the Arabian Peninsula and Southern Sahara, deserve more thorough investigation. Several hypotheses for these disagreement areas are put forward here. Lastly, we comment on the usefulness of the TES instrument for trends analysis and future global observations.

J10.7

Investigating Transported and Local Carbon Monoxide in the Southern Hemisphere with Satellite and Ground-based Remote Sensing

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Distinguishing the relative contribution of transported and local sources of atmospheric pollution is fundamental to developing realistic air quality policies and providing accurate air quality forecasts. We use the different sensitivities of satellite and ground based remote sensing instruments to provide complementary information about sources of carbon monoxide (CO). Total column amounts of CO are compared between the satellite-borne Measurements of Pollution in the Troposphere (MOPITT) and ground-based solar FTIR instruments in the TCCON and NDACC measurement networks. Observations are compared at three Southern Hemisphere stations: Darwin and Wollongong in Australia and Lauder in New Zealand. MOPITT has maximum sensitivity in the free troposphere and measurements are used to interpret long-range transport from continental sources. However, satellite measurements provide limited fine-scale information due to sparse measurement timing and spatial averaging, often missing local pollution events. In contrast, ground-based solar-tracking FTIR instruments have enhanced sensitivity closer to the surface, and can help interpret fine-scale chemistry and dynamic influence. However, FTIR measurements are limited to one location and have trouble interpreting transported signals.

Anomalies in the CO timeseries from each instrument are discussed in relation to pollution delivery pathways of local, regional and long-distance origin. While large-scale pollution events are captured by both instruments, only the satellite instrument can provide regional and global context. For example, the wider geographical impact of Australian fires, such as the severe bushfires around Canberra in 2003, can be traced in the satellite observations and resulting CO plumes tracked out across the Pacific Ocean. MOPITT can also be used to track long-range transport of pollution from biomass burning in South America and southern Africa, reflecting the hemispheric impact of these sources. In comparison, the FTIR can additionally capture local urban and biomass burning influences. Seasonal and interannual variability of CO is significantly different at each site. The roles of emissions and meteorology in CO variability is investigated using global atmospheric modeling with CAM-chem. These modeling studies help quantify the relative impact of local and remote pollution sources to total column CO at the three stations.

J10.8

Trace Gas Measurements and Models in the Mid Atlantic States: Policy Relevant Science

Russell Dickerson, University of Maryland, College Park, MD; and X. Ren, R. J. Salawitch, T. Canty, D. Allen, A. M. Thompson, M. Woodman, and G. T. aburn

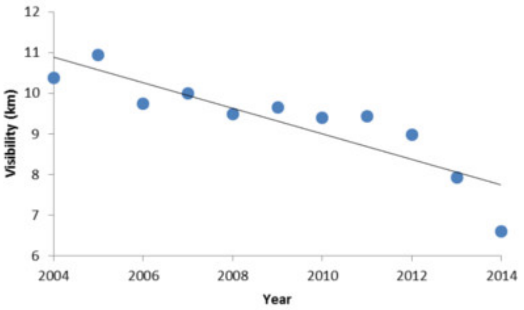
The Mid Atlantic States suffer experience high levels of ozone and particulate matter associated with photochemical smog and haze. Interstate transport as well as the proximity to large water bodies and natural forests complicate the meteorology and chemistry of pollution formation and spatial scales. We describe an integrated approach of in situ and remote sensing to constrain and refine numerical models to better simulate past atmospheric chemistry and future air quality. Findings include the role of the sea and bay breezes, NOx sequestering and release from reservoir species such as isoprene nitrates, and corrections to emissions of key precursors. The results have been used by policy makers to substantially improve air quality in the East and are being extended to studies of greenhouse gas emissions.

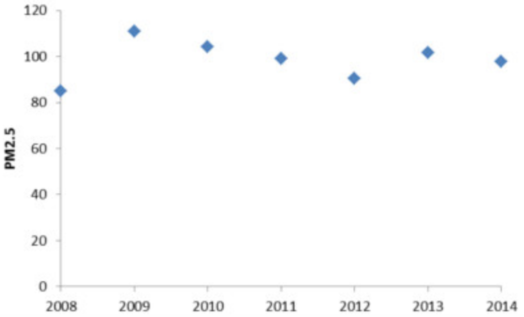
POSTERS

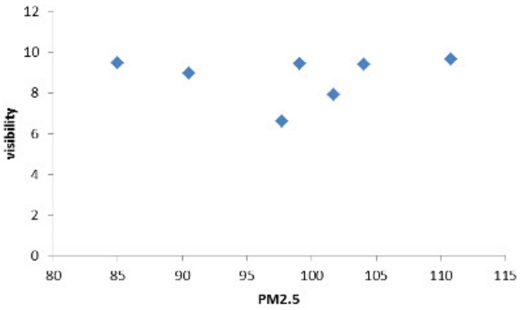
1. Impacts of Aerosols on Precipitation and Thunderstorms in China

Li Cai, Wuhan University, Wuhan, China; and Y. Lin and R. Zhang

As the world's second largest economy, China has experienced severe haze pollution, with fine particulate matter (PM) recently reaching unprecedentedly high levels across many cities. Aerosols interact directly and indirectly with the Earth's radiation budget and climate. For the direct effect, aerosols scatter and absorb solar radiation. Light scattering by aerosols changes the radiative fluxes at the top-of-atmosphere (TOA), at the surface, and within the atmospheric column, while aerosol absorption modifies the atmospheric temperature structure, decreases the solar radiation at the surface, and lowers surface sensible and latent fluxes, suppressing convection and reducing cloud fraction. Furthermore, aerosols indirectly impact climate by altering cloud development, lifetime, precipitation, and albedo, by serving as cloud condensation nuclei (CCN). Here in this paper, we investigate the link between long-term changes in aerosols and precipitation and thunderstorms in China, on the basis of observations and modeling simulations. We illustrate that elevated aerosol loading in China exerts profound impacts on the precipitation patterns and thunderstorm occurrence.







2. Hygroscopic Characteristics of Alkylaminium Carboxylate Aerosols

Mario E. Gomez Hernandez, Florida International University, Miami, FL

The hygroscopic growth factor (HGF) and cloud condensation nuclei (CCN) activity for a series of alkylaminium carboxylate aerosols have been measured using a hygroscopicity tandem differential mobility analyzer coupled to a condensation particle counter and a CCN counter. The particles, consisting of the mixtures of mono- (acetic, propanoic, p-toluic, and cis-pinonic acid) and di-carboxylic (oxalic, succinic, malic, adipic, and azelaic acid) acid with alkylamine (mono-, di-, and tri-methylamines), represent those commonly found under diverse environmental conditions. The hygroscopicity parameter (k) of the alkylaminium carboxylate aerosols was derived from the HGF and CCN results and theoretically calculated. The HGF at 90% RH is in the range of 1.3 to 1.8 for aminium monocarboxylates and 1.1 to 2.2 for aminium dicarboxylates, dependent on the molecular weight and functionality (i.e., the carboxylic or OH functional group in organic acids and methyl substitution in alkylamines). The k value for all aminium carboxylates is in the range of 0.06-1.37 derived from the HGF measurements at 90% RH, 0.05 – 0.49 derived from the CCN measurements, and 0.22-0.66 theoretically calculated. The measured hygroscopicity for the alkylaminium carboxylates increases with decreasing acidity. A deliquescence point is apparent for several of the aminium di-carboxylates, but not for the aminium mono-carboxylates. Our results reveal that alkylaminium carboxylate aerosols exhibit distinct hygroscopic and deliquescent characteristics that are dependent on their molecular functionality, hence regulating their impacts on human health, air quality, and direct and indirect radiative forcing on climate.

3. Reactivity of Ferryl (FeIVO2+) and Mn(III) with Inorganic and Organic Species of Atmospheric Liquid Phases

Virender K. Sharma, Texas A&M University, College Station, TX

It is important to perform studies on the reactions occurring within raindrops or cloud droplets in order to understand the role of heterogeneous chemistry in the atmosphere. Several in-situ measurements have given significant concentrations of transition metals in wet aerosol, cloud, rain, fog, or snow. However, there is still large uncertainly in the understanding of the reactivity of transition metal ions (TMI) in aqueous solution. This creates difficulties in evaluating the impact of cloud chemistry on the atmosphere. Iron (Fe), is the major metal involved in chemical interactions occurring in atmospheric liquid phase and are principally related to the homogeneous aqueous phase chemistry. The production of iron(IV) (ferryl, FeIVO2+) in the environment may be possible through a reaction of Fe(II) with ozone and as an intermediate in the Fenton reaction. Ferryl ion has the potential oxidative role in cloud water. Tthe reactivity and products of reactions of ferryl with constituents of atmospheric droplet will be presented. Assessment of the ferryl oxidative capacity in cloud droplets will be presented. The mechanism and role of the ferryl-substrate system in the atmosphere will be discussed. Manganese also interacts with the chemistry of HO, HO2, and sulfur to influence the oxidizing capacity of the atmospheric aqueous phase. Manganese(II)-Mn(III) redox chemistry plays an important role in the radical chemistry of troposphere by destroying major atmospheric oxidants, O3 and H2O2. Examples of the reactions are: Mn2+ + OH → Mn3+ + OH- and Mn(III) + H2O2 → Mn2+ + HO2 + H+. Manganese(III) generated by such reactions may also react with other constituents of atmospheric water phases. The impact of Mn(III) to the chemistry of atmospheric liquid phase will be given.

4. Atmospheric Aerosol Nonsphericity Over the Loess Plateau in Northwest China

Tian Pengfei, Key Laboratory for Semi-Arid Climate Change of the Ministry of Education, Lanzhou, China

Aerosol nonsphericity, which is not well depicted in model calculations, seriously affects aerosol optical properties and subsequently alters the radiative forcing of the earth-atmosphere system. Based on aerosol backscattering linear depolarization ratio data observed by a polarization lidar at the Semi-Arid Climate and Environment Observatory of Lanzhou University (SACOL) from September 2009 to August 2012 and numerical computations, the spatial and temporal distribution of the aerosol depolarization ratio, parameterization of the derived aspect ratio and influence of water vapor on aerosol nonsphericity were investigated. The aerosol depolarization ratios decreased with increasing height. Aerosol nonsphericity exhibited considerable seasonal variations with a pronounced maximum in spring, when more nonspherical aerosols were transported upward to the free troposphere. The column-averaged lidar depolarizationratios were 0.13, 0.09, 0.08, and 0.10 in the spring, summer, autumn and winter, respectively.Aerosol aspect ratios were derived by combining the lidar observed depolarization ratios and numerical computations. The derived aspect ratios ranged from 1.00 to 1.30, and the frequency distribution was akin to a log-normal distribution that peaked at approximately 1.06. A modified lognormal function was fitted to the frequency distribution of the derived aspect ratios. When the precipitable water was quite small, there was no obvious dependency between the aerosol depolarization ratio and precipitable water. However, in summer, when there was sufficient precipitable water, there was a significant decreasing trend of the depolarization ratio with increasing precipitable water. Moreover, the precipitable water explained 80.8% of the variation in the averaged depolarization ratio in summer.

5. Biomass Burning Smoke and Deep Convection during the 2011 Midlatitude Continental Convective Clouds Experiment (MC3E)

Timothy Logan, University of North Dakota, Grand Forks, ND

Three deep convective cloud cases were selected during the 2011 Mid-Latitude Continental Convective Clouds Experiment (MC3E). Although biomass burning smoke advected from Mexico and Central America was the dominant source of cloud condensation nuclei (CCN) for deep convective cloud formation, the 11 May, 20 May, and 23 May cases exhibited different convective characteristics. The convection in the 11 May and 23 May cases formed in smoke laden environments in the presence of convective available potential energy (CAPE) values exceeding 1000 m2 s-2 and 3000 m2 s-2 along with low-level (0-1 km) shear of 10.3 m s-1 and 5.1 m s-1, respectively. The 11 May case had linear convection while the 23 May case featured discrete supercells. The 20 May case featured elevated linear convection that formed in a more moist environment with cleaner aerosol conditions, weak CAPE (<50 m2 s-2), and stronger low-level shear (25.6 m s-1). Though the 20 May case had the highest precipitation amount and duration, the 23 May case had the highest ice water content (IWC) in the upper levels of the convection (>9 km) suggesting a warm rain suppression mechanism caused by a combination of strong aerosol loading, large CAPE, and weak low-level wind shear. The observed results for the 20 May and 23 May cases agree well with recent modeling studies that simulated the convection and precipitation in these cases. Furthermore, the modeling of the 11 May case is suggested since the abundant amount of smoke CCN did not greatly enhance the overall precipitation amount and could be a possible aerosol-induced precipitation suppression case.

6. Impacts of Megacities on Air Pollution and Climate

Tong Zhu, Peking University, Beijing, China

Megacities are areas with the most intensive human activities, including economic and social activities, accompanied with tremendous energy consumption. These activities lead to concentrated emissions of air pollutants, greenhouse gases, and waste heat, which impact terrestrial and aquatic ecosystems as well as air quality and climate. The negative impact of megacities on local air quality has long been recognized. In recent years, the impact of anthropogenic emissions from megacities on regional and global climate has also received increasing attention. Both of these impacts are linked through energy consumption derived from fossil fuel combustion with emissions that change the atmospheric concentrations of short-lived species that impact both human health and climate, such as aerosols and ozone. Because of these linkages, it has been argued that megacities are the best places to realize the co-benefits of simultaneously controlling air pollution and reducing climate change.

With the growing trend towards urbanization, understanding the role of megacities in local to global atmospheric chemistry is critical to effectively realize the co-benefits of controlling air pollution and reducing climate change. The scientific and engineering knowledge that has been accumulated when developed megacities dealt with their air quality problems in earlier years is a significant resource for current and future megacities. Experiences of developed countries show that the pronounced air quality degradation that accompanied past development can be avoided. In recent years, there have been a growing number of internationally coordinated integrated studies and collaborative projects examining the impacts of megacities on air pollution and climate change. An assessment of these research results and general information about megacities.

7. Optical Properties of Polymers Relevant to Secondary Organic Aerosols

Wilmarie Marrero-Ortiz, Texas A&M University, College Station, TX; and Z. Du, Y. Wang, M. E. Gomez, S. Guo, S. Madronich, M. Hu, and R. Zhang

Aerosols play a critical role in climate directly by scattering and absorbing solar radiation, and indirectly by altering cloud formation; both representing the largest uncertainties in climate predictions. Secondary organic aerosols (SOA), which are produced by chemical reactions and gas-to-particle conversion of volatile organic compounds in the troposphere, represent an important fraction of the total aerosol burden. Some SOA particles are light-absorbing species, known as a Brown Carbon (BrC). The overall contribution of SOA to BrC and the related climate forcing are poorly understood and not currently included in global climate models. This is due in part to the chemical complexity of SOA, and the lack of understanding of SOA formation, transformation, and optical properties. Recent studies indicate the importance of aqueous chemistry in the light-absorbing and high molecular weight oligomeric species, which increase the SOA mass production, and alter the direct and indirect effect of aerosols. In the present study, the multi-phase aerosol chemistry was simulated with bulk phase reactions between small α-dicarbonyl compounds with amines, producing light-absorbing species. Spectroscopic techniques were used to quantify the optical properties and to characterize the product distribution of model SOA formed by these aqueous-phase reactions. Differences between primary, secondary and tertiary amines with glyoxal and methylglyoxal were observed in terms of SOA browning efficiency. Also, an enhancement of the absorption properties was observed at shorter wavelengths. Atmospheric implications of our present work for understanding the formation of light-absorbing SOA will be presented.

8. The Kinetics of the ClO-Dimer Cycle

Ross J. Salawitch, University of Maryland, College Park, MD; and T. Canty

Molina and Molina (JPC, 1987) first proposed that the ClO-dimer cycle would be an important route for the catalytic removal of stratospheric ozone. Here, we use a photochemical model of halogen chemistry, constrained by in-situ aircraft observations of [ClO] and [ClOOCl], to test our understanding of the kinetics of this cycle. We will show simulations of daytime observations for a variety of laboratory determinations of the rate constant for formation of ClOOCl via the self-reaction of ClO as well as for the absorption cross section of ClOOCl (σ_{ClOOCl}). Nighttime observations of [ClO] and [ClOOCl] will be compared to simulations based on various laboratory and theoretical determinations of the ClO/ClOOCl equilibrium constant. Our analysis suggests ClOOCl photolyzes more quickly than inferred from the most recent laboratory determination of the σ_{ClOOCl} (Young et al., GRL, 2014), which is the first study to span all wavelengths relevant for atmospheric photolysis. We also show gaps remain in our understanding of the nighttime partitioning of [ClO] and [ClOOCl], for cold conditions typical of the polar lower stratosphere during winter.

9. Variation of Radiative Properties During Black Carbon Aging: Theoretical and Experimental Intercomparison

Cenlin He, University of California Los Angeles, Los Angeles, CA; and K. N. Liou, Y. Takano, R. Zhang, M. Ievy, P. Yang, Q. Li, and L. R. Leung

A theoretical black carbon (BC) aging model is developed to account for three typical evolution stages, namely, freshly emitted aggregates, coated BC by soluble material, and BC particles undergoing further hygroscopic growth. The geometric-optics surface-wave (GOS) approach is employed to compute the BC single-scattering properties at each aging stage, which are subsequently compared with laboratory measurements. Theoretical calculations are consistent with measurements in extinction and absorption cross sections for fresh BC aggregates, but overestimate the scattering cross sections for BC mobility diameters of 155, 245, and 320 nm, because of uncertainties associated with theoretical calculations for small particles as well as laboratory scattering measurements. The measured optical cross sections for coated BC by sulfuric acid and for those undergoing further hygroscopic growth are captured by theoretical calculations using a concentric core-shell structure, with differences of less than 20%. This suggests that the core-shell shape represents the realistic BC coating morphology reasonably well in this case, which is consistent with the observed strong structure compaction during aging. We find that the absorption and scattering properties of fresh BC aggregates vary by up to 60% due to uncertainty in the BC refractive index, which, however, is a factor of two smaller in the case of coated BC particles. Sensitivity analyses on the BC morphology show that the optical properties of fresh BC aggregates are more sensitive to fractal dimension than primary spherule size. The absorption and scattering cross sections of coated BC particles vary by more than a factor of two due to different coating structures. We find an increase of 20-250% in absorption and a factor of 3-15 in scattering during aging, significantly depending on coating morphology and aging stages. Applying the aging model to CalNex 2010 field measurements, we show that the resulting BC direct radiative forcing (DRF) first increases from 1.5 to 1.7 W m-2 and subsequently decreases to 1.0 W m-2 during the transport from the Los Angeles Basin to downwind regions, as a result of the competition between absorption enhancement due to coating and dilution of BC concentration. The BC DRF can vary by up to a factor of two due to differences in BC coating morphology. Thus, an accurate estimate of BC DRF requires the incorporation of a dynamic BC aging process that accounts for realistic morphology in climate models, particularly for the regional analysis with high atmospheric heterogeneity.

10. Global Climate Models Intercomparison of Anthropogenic Aerosols Effects on Regional Climate over North Pacific

Jiaxi Hu, Texas A&M University, College Station, TX; and Y. Wang, Y. Lin, B. Pan, and R. Zhang

Aerosols can alter cloud microphysics and macrophysics, which may further impact weather and global climate. With the development and industrialization of major Asian countries, anthropogenic aerosols have attracted considerable concerns and remain to be the largest uncertainty to estimates and interpretations of Earth's changing energy budget. Here we assess the performance of two stat-of-art global climate models (National Center for Atmospheric Research-Community Atmosphere Model 5 (CAM5) and Geophysical Fluid Dynamics Laboratory Atmosphere Model 3 (AM3)) by the impacts of anthropogenic aerosols on Pacific storm track region (30N – 50N, 120E -120W). Simulations are based on two aerosol scenarios, i.e. present day (PD) and pre-industrial (PI), to exhibit the long-range transport of anthropogenic aerosols across north Pacific. Both models show aerosol optical depth enhanced (AOD) by around 22%, with CAM5 AOD 40% lower in magnitude. Ice water path (IWP), stratiform precipitation, convergence and vertical velocity change in the two models show different trend and magnitude. AM3 generally shows qualitatively good agreement with long period satellite observations while CAM5 overestimates convection and liquid water path resulting in an underestimation of large-scale precipitation and IWP. Due to coarse resolution and parameterization in convection schemes, both models' performance on convection needs to be modified. Aerosols performance on large-scale circulation and radiative budget are also examined in this study.

11. Sensitivity of Contribution of Organic Aerosol on Aerosol Optical Properties in Refractive Index and Hygroscopicity

Chang Hoon Jung, Kyungin Women's University, Incheon, South Korea; and J. Lee, H. J. Shin, and Y. P. Kim

In this study, the sensitivity of organic aerosol on aerosol optical properties is conducted. Both the refractive index of Organic aerosol as well as hygroscopicity was considered. Based on the filter based monthly averaged sampling measurement data at the intensive observation sites in Seoul, Korea, the contribution of each composition on the aerosol optical properties were estimated. The aerosol optical properties are simulated by combining aerosol dynamic model for polydispersed aerosol with optical properties model using Mie theory. The obtained optical properties such as extinction, scattering and absorption coefficient were calculated AERONET Aerosol Optical Thickness measurement data. In order to estimate contribution of the light absorption and hygroscopicity properties of Organic Aerosol on the aerosol optical properties, the sensitivity test was conducted with different imaginary refractive index and organic aerosol hygroscopic growth factor. The results show that mass extinction and absorption efficiency increases linearly as imaginary refractive index increase, which means one can estimate the mass absorption efficiency of organic aerosol using the linear regression as a function of imaginary refractive index. The results also show that mass extinction and absorption efficiency decreases as hygroscopic factor of organic aerosol increases. The contribution of organic aerosol on the mass extinction efficiency, however, depends on the chemical composition of other aerosol mixture.

12. Assessment of the Impacted Area Generated by Freight Transportation over Unpaved Roads Using CFD

Jose Huertas, Instituto Tecnologico y de Estudios Superiores de Monterrey, Toluca, Estado de Mexico, Mexico; and D. Prato Sr., J. C. Miranda Sr., and A. Valencia Sr.

The passing of vehicles cause air pollution problems due to particulate matter (PM) around the unpaved roads. By regulatory purposes, the environmental authorities need to know the extent of the affected area. Looking to address this need, the dispersion of PM was modeled around unpaved roads using computational fluid dynamics (CFD). The model was validated with the experimental measurements results of concentration of PM made in the perpendicular direction of two unpaved roads located on level ground without additional sources of PM. It was found that the results obtained with the model are correlated (R2 <0.77) with experimental measurements and the model underestimates the concentration values of PM. The distance at which the particles are deposited increases with the wind speed until a speed of 3.5 m/s. At this distance, the deposition speed becomes constant. It was also found that the concentration profile of PM with the distance to the road can be expressed in generalized form using three dimensionless numbers. Taking advantage of the generic nature of the results, a methodology was developed to estimate the average annual concentration of PM modeling successive short intervals of time where we can assume a steady-state phenomenon. This alternative substantially reduce the computation time. Finally, the extension of the impacted area and the distance at which the concentration profile equals the maximum concentration values of PM of NAAQS was defined. This distance was 93 and 147 m for roads where experimental work was performed.

13. Methodology to Model the Dispersion Pollutants Exposed to Chemical Reactions, Using Gaussian Models

Jose Huertas, Instituto Tecnologico y de Estudios Superiores de Monterrey, Toluca, Estado de Mexico, Mexico; and D. Prato and A. Valencia Sr.

The environmental impacts caused by the operation of gas-burning equipment are represented by high levels of surface level concentration of air pollutants (criteria pollutants and toxic).
It has become a need to find the optimun operation condition of most common applications of thermal combustion equipment so that do not imply a risk on the surrounding areas.
AERMOD was used to model the dispersion of pollutants generated by burning waste gas with typical composition in Colombia. It has consider the case of an emission source of generic characteristics, located in a flat surface and under general weather conditions (multiple Meteorology).
A sensitivity analysis of the critical pollutants concentration, subject to changes in the input parameters was developed, such as concentration levels of those pollutant in the emission source, gas flared volume, etc. This information are used to find the optimal operational conditions of gas flaring that lead to avoid the risk in the concentration levels of the pollutants in the surrounding areas.
In this study it was found that the concentration of SO2 at surface level is highly sensitive to the content of H2S and the volumetric flow of gas flaring (YSO2 Q = 0.0155 for the 5th up to 3 hr). in a less significant way, the height of the chimney (YSO2 = -11 334 Ln (h) + 68923, for up to 3 hr the 5th) and very insensitive to the output speed of the gases in the chimney.

14. Soot Formation and Properties from Combustion of Toluene in a Shock Tube

Chong Qiu, University of North Alabama, FLORENCE, AL; and A. F. Khalizov, R. Zhang, B. Hogan, and E. L. Petersen

Aromatic organic compounds are commonly found in various fossil fuels and their combustion in engines produces soot that has significant impacts on air quality, human health, and climate. Using toluene as a proxy, we have investigated the formation and properties of soot particles from the combustion of aromatic compounds in a shock tube. Several important properties of the soot particles were measured, including size distributions, effective density, elemental carbon (EC) to organic carbon (OC) ratio, morphology, and optical properties (light absorption, scattering, and single scattering albedo). The dependence of soot properties on combustion temperature and fuel equivalent ratio was explored. Combustion temperature showed significant impacts on particle size distribution, morphology, and EC/OC ratio. In general, higher combustion temperature yielded larger, more fractal soot particles with higher EC content. Soot yield increased as the fuel equivalent ratio increased. Increases in combustion temperature and fuel equivalent ratio enhanced the aerosol light absorption and reduced the single scattering albedo. Compared to propane and diesel fuel, toluene combusted in the shock tube produced significantly more soot at a faster rate and with an earlier soot inception. Our results contribute to a better understanding of the soot formation mechanism and properties, helping to assess climate and environmental impacts from the combustion of fossil fuels.

15. Radiative Properties of Soot Superaggregates in the Infrared

Yang Yu, Aerosol Impacts & Research (AIR) Laboratory, St. Louis, MO; and I. Arnold and R. K. Chakrabarty

Soot is one of the largest man-made contributors to global warming and its influence on climate has been greatly underestimated. This underestimation is due to poor understanding of the microphysical properties of soot and its parameterization in climate models and satellite retrieval algorithms. Very recently, researchers observed a new kind of soot particle “super aggregates”, emitted from large-scale forest fires globally. The morphology of these particles is described by a mass fractal dimension ≈ 2.6, mobility diameter > 1 μm, and aerodynamic diameter in the range of 0.5 - 20 μm. Given their large size, it is expected that these soot superaggregates would impact radiative forcing in the longer wavelengths (i.e. infrared (IR)) of the incoming solar spectrum. Here, we quantitatively investigate the microphysical and optical properties of soot superaggregates using controlled laboratory-scale experiments. Soot superaggregates were produced in our laboratory using a novel diffusion flame aerosol reactor operated in a negative gravity. Next, an Integrated Photoacoustic-Nephelometer (IPN) System using a 1047nm laser source was built to simultaneously measure absorption and scattering coefficient of these particles in real-time. Along with the optical properties, the total number concentration and aerodynamic size distribution were measured using a TSI Aerodynamic Particle Sizer. The mobility size distribution was also inferred from electron microscopy analysis of a statistically significant number of particles. Our results show that soot superaggregates have distinct physical and optical properties compared to conventional sub-micron soot aggregates, and could significantly impact direct radiative forcing in the atmosphere.

16. Impacts of Saharan Dust on Regional Climate and Tropical Cyclogenesis over the Atlantic Basin

Bowen Pan, Texas A&M University, College Station, College Station, TX; and Y. Wang, J. Hu, Y. Lin, and R. Zhang

Saharan dust can exert substantial radiative and microphysical effects on the regional climate system. Its impacts on the genesis and intensification of tropical cyclones (TCs) remain unclear. In this research, the influences of Saharan dust on the Atlantic regional climate and the genesis of TCs are investigated in the hurricane seasons of 2005 and 2006, which represent the active and inactive hurricane seasons respectively. The atmospheric stand-alone version of the Community Earth System Model version 1.0.4 (CESM1.0.4), CAM5.1, is used to simulate the climate condition in full (dust) and none dust (non-dust) scenarios. Two regions of interest, the Atlantic TC genesis region (GNR, 50W-20W, 5N-15N) and the TC intensification region (ITR, 70W-40W, 15N-30N), are defined by Lau and Kim [2007] and are investigated. Model output proves the important impacts of the Saharan dust on the radiative budget, hydrological cycle, and TC genesis. The dust perturbs the large-scale circulation that moves the ITCZ northward, enhances the West African monsoon, changes the cloud fraction, and perturbs the regional longwave and shortwave radiations. Dust favors the genesis of TCs thermodynamically by increasing the mid-level moisture in the GNR but suppresses the TC formation by increasing the vertical wind shear and decreasing low-level vorticity in the GNR. It is likely that the TC genesis region shifts northward with the ITCZ.

17. Acid-catalyzed Multiphase reaction of Oxygenated Volatile Organic Compounds with Hydrogen Peroxide

Weigang Wang, Institute of Chemistry, Chinese Academy of Sciences, Beijing, China; and Q. Liu and M. Ge

The acid-catalyzed multiphase reaction with hydrogen peroxide has been suggested to be a potential pathway for secondary organic aerosol formation from isoprene and its oxidation products. However, knowledge of the chemical mechanism and kinetics for this process is still incomplete. Herein the uptake of several oxygenated volatile organic compounds on H2SO4-H2O2 mixed solutions was investigated using a flow-tube reactor coupled to a mass spectrometer. These OVOCs, including methacrolein, 2-methyl-3-buten-2-ol (MBO232), 2-methyl-2-butanol, 3-buten-2-ol, 2-butanol and 3-methyl-2-buten-1-ol (MBO321) are related to isoprene photooxidation or structurally similar to isoprene. The reactive uptake coefficients were acquired and the reaction pathways were proposed according to the products information.

Isoprene was generated from the reaction of MBO232 and H2SO4. When adding H2O2 into the acid solutions, new peaks were detected by the mass spectrometer, indicating that a different reaction mechanism

occurred in H2SO4-H2O2 mixed solution compared to that in H2SO4 solution only. To further survey this chemical mechanism, the uptake of three aliphatic alcohols (2-methyl-2-butanol, 3-buten-2-ol, 2-butanol) on H2SO4-H2O2 mixed solution was studied. Tertiary or allyl alcohols can contribute to organic hydroperoxides (ROOHs) formation but secondary alcohols cannot under through this path according to the experimental results of gas-liquid multiphase reaction. The generation and degradation mechanisms of ROOHs were proposed using various methods, such as isotope labeling, infrared characterization and mass spectrum measurements. Once formed, ROOHs were found to undergo two degradation pathways: the acid-catalyzed rearrangement reaction and the organic hydrogen peroxysulfate formation pathway. As for the MBO321-H2SO4/ H2O2 system, ROOHs were also found in the gas and liquid phase through the acid-catalyzed route, while the formation of acetone and acetaldehyde were also detected which could undergo a rearrangement reaction. Organosulfates, which were proposed to be SOA tracer compounds in the atmosphere, were also produced during the oxidation process. These results indicate that acid-catalyzed multiphase oxidation of typical OVOCs with hydrogen peroxide may contribute to SOA mass under certain atmospheric conditions.

18. Measurements of Diffusion Coefficients of Organics Dyes in Proxies of Atmospheric Particles and Comparison with Predictions Using the Stokes-Einstein Equation

Dagny Anna Ullmann, University of British Columbia, Canada, Vancouver, British Columbia, Canada; and Y. Chenyakin, S. Kamal, and A. K. Bertram

Because of their high impact on the environment, particles consisting of secondary organic material (SOM) have moved into the centre of attention in atmospheric research. These SOM particles are formed in the atmosphere and can influence the Earth's climate directly and indirectly by absorption and scattering of solar radiation and acting as ice and cloud nuclei. Furthermore, it has been found that SOM particles can affect the human health and reduce air quality e.g. visibility significantly.

To better understand the effect of SOM particles on the Earth's climate, several characteristics of SOM particles of different compositions have been studied. Diffusion rates are essential for the investigation of processes such as particle growth and the rate of heterogeneous reactions within SOM particles. In the past, diffusion rates of SOM particles have been estimated by converting viscosity values into diffusion coefficients using the Stokes-Einstein relation.

Here, we present values of diffusion coefficients, which were measured directly using the fluorescence microscopy method FRAP (fluorescence recovery after photobleaching), of organic dyes in organic-water proxies of atmospheric particles. Fluorescent dyes of varying sizes and molecular weights (320-1.50×10^5 g/mol) were used and diffusion coefficients were measured across a wide range of relative humidities (40-93% RH). These measurements add to the limited amount of data on the diffusion rate constants of organics within organic-water solutions.

19. Aerosol Effects on Precipitation and Lightning Activity over Houston, Texas

Yun Lin, Texas A&M University, College Station, TX; and Y. Wang and R. Zhang

Cloud-to-ground lightning flash from the National Lightning Detection Network (NLDN) in Houston, Texas shows a higher density over the urban area based on a twelve-year period (1989-2000) analysis. The local enhancement of lightning centered at Houston urban region could be attributed to the urban heat island effect and anthropogenic pollution. A two-moment bulk microphysical scheme has been implemented into the Weather Research and Forecasting (WRF) model to investigate urban aerosol effects on lightning associated with a typical sea breeze frontal event occurring in Houston. The simulated radar reflectivity generally agrees with the observations and the calculated lightning potential index (LPI) exhibits temporal and spatial consistence with lightning flashes recorded by NLDN. Sensitivity study indicates that aerosols greatly enhance lightning activity under the polluted condition. The analysis on the microphysical properties documents that more efficient mixed phase processes and intensified convection under the polluted aerosol condition lead to more efficient graupel-ice collision, which favors electrification and charge separation in thunderstorms. In addition, aerosols suppress the light precipitation process but increase the heavy rain during the studied sea breeze case. The aerosol radiative effects on lightning activity will be examined in the next step through incorporating the aerosol optical properties in the radiative scheme of the model. Urban heat island effect will be investigated in near future as well.

20. Decadal Record of Ammonia Observed by AIRS

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Ammonia is an integral part of the nitrogen cycle and is projected to be the largest single contributor to each of acidification, eutrophication and secondary particulate matter in Europe by 2020 (Sutton et al., 2008). The impacts of NH3 also include: aerosol production affecting global radiative forcing, increases in emissions of the greenhouse gases nitrous oxide (N2O) and methane (CH4), and modification of the transport and deposition patterns of SO2 and NOx. Therefore, monitoring NH3 global distribution of sources is vitally important to human health with respect to both air and water quality and climate change. We have developed new daily and global ammonia (NH3) products from AIRS hyperspectral measurements. These products add value to AIRS's existing products that have made significant contributions to weather forecasts, climate studies, and air quality monitoring. With longer than 13 years of data records, these measurements have been used not only for daily monitoring purposes but also for inter-annual variability and short-term trend studies. We will discuss the global NH3 emission sources from biogenic and anthropogenic activities over many emission regions captured by AIRS. We will focus their variability in the last 13 years. Validation examples using in situ measurements for AIRS NH3 will also be presented.

21. Anthropogenic Influence on Decadal Aerosol Trends over the Western North Atlantic Ocean

Andrew Jongeward, University of Maryland, College Park, MD; and Z. Li and J. Xiong

Aerosols from both natural and anthropogenic sources contribute to atmospheric variability and impact Earth's radiative balance around the globe. Atmospheric aerosols can also indirectly influence the radiative balance through interactions with clouds. Industry and other human activities during the last century have led to an increase in the release of trace gases and anthropogenic aerosol particles. More recently, policies aimed at reducing air pollutants for human health concerns have seen success at improving local air quality. Regional anthropogenic contribution to the atmospheric aerosol loading is presumed to be altered as a by-product.

In this work we examine variations in aerosol loading over the eastern seaboard of the United States and the North Atlantic Ocean from 2000 to 2012. This period is significant due to anthropogenic emissions control measures that are in place. We use monthly mean data from satellite (MODIS), ground (AERONET, IMPROVE), and model (GOCART) sources. From MODIS observations, a statistically significant -0.020 decade^{-1} trend in aerosol optical depth (AOD) over the mid-latitudes is seen. Correlation of AOD between MODIS and AERONET sites in the upwind regions as well as trend analysis of GOCART component AOD reveal the anthropogenic origin of this trend downwind of the eastern U.S. IMPROVE network surface $PM_{2.5}$ observations from the eastern U.S. confirm statistically significant decreases in total $PM_{2.5}$ driven primarily by decreases in anthropogenic $PM_{2.5}$ species. In contrast to the mid-latitudes, MODIS observations reveal a statistically significant 0.015 decade^{-1} trend in AOD over the sub-tropics. AERONET correlation, GOCART component trends, and an estimation of summertime dust flux suggest that this trend is of natural origin. Finally, preliminary results are presented for both aerosol direct and indirect effects. This work offers an assessment of aerosol trends and aerosol-cloud interactions in the context of decreasing anthropogenic aerosol contribution and on timescales relevant to climate processes.

22. Size-differentiated Inorganic Chemical Composition of Atmospheric Particles at the T1 Supersite during the MILAGRO Field Campaign

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Cascade impactors with eight cut stages were used to sample atmospheric particles at Tecámac (T1 supersite), State of Mexico, during the MILAGRO field campaign in March 2006. The sampling site is located in the northeast (NE) edge of Mexico City; it experiences both fresh local emission and aged pollutants produced in Mexico City. Samples were analyzed to determine total mass, Ca^{2+}, Na^{+}, Mg^{2+}, NH_4^{+}, K^{+}, Cl^{-}, SO_4^{2-}, and NO_3^{-} concentrations. Mexico City pollution plume contributes with particle mass in all aerodynamic diameter sizes, but local emissions are quite important. PM_{10} average mass concentration was 30.3 µg m^{-3}; for coarse mode (3.2 – 10 µm) was 11.7 µg m^{-3} and fine mode (0.18 – 1.8 µm) was 18.6 µg m^{-3}. The ions NO_3^{-}, SO_4^{2-}, Na^{+}, and NH_4^{+} were found in high concentrations on fine mode, and high concentrations of Cl^{-}, K^{+}, Ca^{2+}, and Mg^{2+} were found on coarse mode.

23. A Laboratory Study to Constrain Gas-Phase Formic Acid and Acetic Acid Sources

Dhruv Mitroo, Washington University in St. Louis, St. Louis, MO; and M. Baasandorj, D. B. Millet, and B. Williams

Organic acids contribute to cloudwater and aerosol acidity and are therefore consequential for aqueous phase chemistry in the atmosphere. However, the sources and sinks of the two most abundant organic acids in the atmosphere, formic acid (FA) and acetic acid (AA), are not well characterized. Recent field measurements performed in a range of environments have demonstrated that atmospheric boundary layer gas-phase FA and AA levels exceed modeled levels by up to a factor of 3 (Millet *et al.*, 2015). The gap between modeled and observed FA and AA suggest a substantial and unknown source. What is unclear is whether this source results from poor modeling of volatile organic compounds (VOCs) oxidation chemistry (secondary source), from the oxidation of condensed-phase organic material (secondary source), or an unaccounted flux to the atmosphere (primary source).

We present a series of laboratory studies to assess gas-phase yields of FA and AA from different primary and secondary sources to provide insight in constraining potential contributions from a range of source types and aging processes. We employ a custom built combustion and emission chamber for primary source generation. In addition, a Potential Aerosol Mass (PAM) reactor (Kang *et al.*, 2007) is used to generate secondary sources (i.e., gas-phase photochemical production and production from the oxidative aging of primary particles). The PAM reactor can achieve oxidant exposures equivalent to several days in the atmosphere, thus allowing us to track the gas-phase FA and AA production as a function of precursor type, precursor concentration, oxidative exposure, and oxidant type (e.g., OH vs. O_3).

Detection of gas-phase FA and AA is achieved via a proton-transfer reaction mass spectrometer (PTR-MS; Ionicon Analytik). A custom calibration system is integrated with the PTR-MS to eliminate interfering compounds at selected *m*/*z*'s, as well as quantifying the FA and AA mixing ratios (Baasandorj *et al.*, 2015). Experiments on single-component precursor VOC oxidation in the PAM reactor allows a determination of acid yields from selected compounds that are found in large abundance in the atmosphere (e.g., isoprene and monoterpenes in forested regions, toluene and xylene in urban regions, etc.). Direct primary FA and AA emissions from biomass combustion sources are also characterized. Here, the vapor-phase fraction of biomass combustion emissions are removed using a denuder, and the particle-phase fraction undergoes further aging in the PAM reactor, releasing gas-phase organic acids. A scanning mobility particle sizer (SMPS; TSI Inc.) and an aerosol mass spectrometer (AMS; Aerodyne Research Inc.) are used to quantify the total particulate matter and organic aerosol mass concentrations. Gas-phase acid yields from the oxidative aging of biomass combustion aerosol will be presented. Results from this study can be used to improve our modeling of the sources and sinks of these abundant acids.

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24. Comparing and Verifying WRF Simulations of Water Vapor for TC Ingrid (September 2013)

Thomas Daniel Allison, Tallahassee, FL; and H. E. Fuelberg

Atmospheric water vapor is a potent greenhouse gas. Variations of water vapor in the upper troposphere and lower stratosphere (UTLS) are important in determining climate impacts. However, the water vapor budget of tropical cyclones (TCs) and its impact on the UTLS remain under-studied. Therefore, this paper will describe high-resolution simulations from the Weather Research and Forecasting (WRF) model version 3.6 with finest grid spacing of 1.33 km to calculate the water vapor budget of TC Ingrid during September 2013. After extensive testing on the 12 km domain, the WRF Double Moment 6-class Scheme and the New Simplified Arakawa-Schubert Scheme are chosen for microphysics and cumulus parameterization, respectively. The cumulus parameterization scheme is incorporated only on the coarsest 12 km domain, with the 4 km and 1.33 km domains simulating convection explicitly.

Our WRF simulations of TC Ingrid closely correspond to the National Hurricane Center's (NHC's) best track data. Thus, model results of the water budget are compared to the in situ airborne data of Ingrid collected during NASA's Studies of Emissions and Atmospheric Composition, Clouds, and Climate Coupling through Regional Surveys (SEAC4RS). Comparisons between ER-2 in situ data in the UTLS of TC Ingrid from 13-16 September are used to further verify the WRF model runs. In addition, a variety of satellite imagery is used to validate the structure of TC Ingrid. Time series of water vapor depict spatial and temporal changes of water vapor in the UTLS throughout Ingrid's lifecycle. The paper will describe the physical processes contributing to these changes using the model output.

Since TC Ingrid is a relatively weak and short-lived TC, with maximum sustained winds of category one intensity on the Saffir-Simpson scale, a stronger and longer-lived TC also is modeled in WRF for comparison. Similar options within WRF are used to simulate the water budget of the stronger TC, with results compared to those of TC Ingrid. This paper will benefit the scientific community by showing whether and how TCs hydrate or dehydrate the UTLS, helping to answer a question posed during the SEAC4RS mission. The paper highlights the mechanisms by which water vapor is injected into the UTLS, and the widespread horizontal and vertical transports of water vapor within TCs of varying intensities.

25. OH-Initiated Oxidation of m-Xylene on Black Carbon Aging

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M. Hu, Y. Lin, M. E. Gomez, M. levy, D. R. Collins, and R. Zhang

Laboratory experiments are conducted to investigate aging of size-classified black carbon (BC) particles from OH-initiated oxidation of *m*-xylene. The variations in the particle size, mass, effective density, morphology, optical properties, hygroscopicity, and activation as cloud condensation nuclei (CCN) are simultaneously measured by a suite of aerosol instruments, when BC particles produced from incomplete combustion of propane are exposed to the oxidation products of the OH-*m*-xylene reactions. The BC aging process is governed by the coating thickness (Δrve), which is correlated to the reaction time and initial concentrations ofm-xylene and NOx. For an initial diameter of 100 nm and Δrve = 44 nm, the particle size and mass increase by a factor of 1.5 and 10.4, respectively, and the effective density increases from 0.43 to 1.45 g cm^{-3} due to the organic coating and shrinking of the BC core. The BC particles are fully converted from a highly fractal to nearly spherical form for a Δrve value of 30 nm, as reflected by a decreased BC core size and the dynamic shape factor. The scattering, absorption cross-section, and single scattering albedo are enhanced accordingly. The critical supersaturation for CCN activation is reduced to 0.1%, with an initial particle size of 100 nm and Δrve = 44 nm. Our results imply that the oxidation of *m*-xylene exhibits larger impacts in modifying the BC particle properties than those for the OH-initiated oxidation of isoprene and toluene.

26. Volatile Organic Compounds in the Atmosphere of Mexico City

JESSICA P. Garzon-Barrera, UNIVERSIDAD TECNOLOGICA EQUINOCCIAL DE ECUADOR, Santo Domingo, Santo Domingo, Ecuador; and J. Huertas, M. Huertas, M. Magaña, B. Cardenas, T. Watanabe, S. wakamatsu, S. Blanco-Jimenez, and T. Maeda

64 VOCs, including 36 toxic VOCs, were measured at four sites in the Mexico City Metropolitan Area (MCMA) during 2011-2012. Results showed that Iztapalapa-area presented the highest VOC concentrations, and that VOC compounds related to LPG leakages exhibited the highest concentrations. Acetone and ethanol were the toxic VOCs that showed the highest average concentration (31.9±4.6, 30.8±3.9 ppb). Correlation analysis among measured VOCs indicates that they come from vehicular emissions and industrial sources. Compared to the former field measurement campaign of 2005-2006, the ozone formation potential (OFP) has been reduced by 53%. Based on these results, we propose as alternatives to control ground-level ozone formation in the MCMA to limit the use of oil-based paints, and to restrict the use of vehicle model years prior to 1991, which still use carburetors.

27. Aerosol Hygroscopic Properties in Seoul during the KORUS-AQ Pre-Campaign

Minsu Park, Yonsei University, Seoul, South Korea; and S. S. Yum, N. Kim, G. N. Bae, K. H. Kwak, H. J. Shin, J. S. Park, I. H. Song, and J. Y. Ahn

The prediction of the effects of atmospheric aerosols on human health and climate has been known to have large uncertainty. To reduce the uncertainty about the aerosol effect, many studies have been carried out, but still they are insufficient to cover the whole world. Because Seoul is a megacity located downwind of China, there are abundant anthropogenic sources and transported aerosols in Seoul. KORUS-AQ is initiated as a collaborative field measurement project between Korea and U.S. to investigate formation of ozone and aerosol and interactions between chemistries, transport, and various sources in Seoul. The KORUS-AQ campaign will be held in May to June, 2016, and the pre-campaign for KORUS-AQ was held in May to June, 2015. Here we present the results from the pre-campaign. Submicron aerosol number concentration (N_{CN}), TSI CPC and size distribution (14 nm < particle diameter < 670 nm), TSI SMPS were measured at Korea Institute of Science and Technology (KIST) in Seoul for about four weeks (18 May – 12 June). CCN number concentration (N_{CCN}), DMT CCN counter, aerosol hygroscopic growth factor (H-TDMA), and aerosol chemical composition (Aerodyne AMS) were also measured at the same location. The average diurnal variation of N_{CN} shows that distinct peaks are located at morning rush hours (0600 – 0900) and about 1500, the latter of which indicates that particle formation events frequently occurred in Seoul during the pre-campaign period. However, N_{CCN} does not show noticeable diurnal variation during this period. According to H-TDMA measurement, the fraction of hygroscopic aerosols varied diurnally with the maximum in the afternoon and the minimum during the morning rush hours. For 100 nm particles and larger, most observations showed a bimodal hygroscopic behavior except in the afternoon (1200 – 1800), when unimodal behavior was predominant. For 50 nm particles and smaller, on the other hand, most observation showed a unimodal behavior except during the rush hours. Aerosol chemical composition and its relationship with CCN activity and hygroscopicity will be analyzed and presented at the conference.

28. Polymer Formation on Atmospheric Nanoparticles and Its Effect on Aerosol Hygroscopicity

Wen Xu, Aerodyne Research, Inc, Billerica, MA; and M. Gomez-Hernandez, S. Guo, M. levy,J. R. Secrest, W. Marrero-Ortiz, D. R. Collins, and R. Zhang

Although the dominant source of global aerosol production is new particle formation, the growth mechanisms for the freshly nucleated nanoparticles remain largely uncertain. In the present study, we observed large size growth, dependent on relative humidity and particle size, when 4-20 nm sulfuric acid nanoparticles are exposed to epoxide vapors. After mass spectrometric chemical analysis of the size-enlarged particles, high molecular weight organosulfate and polymer products are revealed. Based on the large formation yield of epoxides from photochemical oxidation of biogenic volatile organic compounds (VOCs), they could play an important role in the growth of freshly nucleated nanoparticles. To investigate the effect of polymers on aerosol hygroscopicity, we used surrogates to mimic atmospherically relevant polymers, including glyoxal trimer dihydrate, methylglyoxal trimer dihydrate, sucrose, and organic-sulfuric acid mixtures. Low κ values were observed for polymers than monomers. Our results indicate that polymer formation decrease hygroscopicity and CCN activity of particles and provide guidance in analyzing ambient aerosols.

29. Impact of Asian Dust Storm on Aerosol Chemistry of Xian, Central China: Rapid Heterogeneous Formation of Nitrate

Gehui Wang, Chinese Academy of Sciences, Xi'an, Shaan Xi, China

Composition and size distribution of atmospheric aerosols from Xi'an city (~400 m, altitude) in inland China during the spring of 2009 including a massive dust event on 24 April were measured and compared with a parallel measurement at the summit (2060 m, altitude) of Mt. Hua, an alpine site nearby Xi'an city. EC (elemental carbon), OC (organic carbon) and major ions in the city were 2–22 times higher than those on the mountaintop during the whole sampling period. Compared to that in the non-dust period a sharp increase in OC was observed at both sites during the dust period, which was mainly caused by an input of biogenic organics from the Gobi desert. However, adsorption/heterogeneous reaction of gaseous organics with dust was another important source of OC in the urban, contributing 22% of OC in the dust event. In contrast to the mountain atmosphere where fine particles were less acidic when dust was present, the urban fine particles became more acidic in the dust event than in the non-dust event, mainly due to enhanced heterogeneous formation of nitrate and diluted NH3. Cl- and NO-3 in the urban air during the dust event significantly shifted toward coarse particles. Such redistributions were further pronounced on the mountaintop when dust was present, resulting in both ions almost entirely staying in coarse particles. On the contrary, no significant spatial difference in size distribution of SO42- was found between the urban ground surface and the mountain atmosphere, which dominated in the fine mode (<2.1 µm) during the nonevent and comparably distributed in the fine (<2.1 µm) and coarse (>2.1 µm) modes during the dust event. Our results also showed that around 90% (in mass) of SO42- during the dust storm event was derived from Gobi desert surface soil, while around 70% of nitrate in the event was derived from heterogeneous reactions of local pollutants (i.e., gaseous HON3 and NOx) with dust particles, indicating a rapid formation of nitrate.

30. Variations of Aerosol Particles Hygroscopicity During the Dust and Haze Events from Filed Measurements in Spring 2015 in Urban Beijing, China

Wang Yuying Sr., Beijing Normal Univerisity, Beijing, China, China; and Z. fang Jr., L. Zhanqing, and L. Ping

Hygroscopicity of atmospheric aerosols has significant impact on the aerosol size distribution, cloud condensation nuclei formation, climate forcing, etc. In the spring of 2015, a Volatility/Hygroscopicity-Tandem Differential Mobility Analyzer (V/H-TDMA) was used to evaluate the hygroscopic properties of aerosol during dust and haze events in urban Beijing. The instrument can also measure the particles number concentration and the volatility with particle size range of 10-400nm. Our preliminary results showed that the diurnal variation of particle number concentration was with double peaks in the morning and in the afternoon of the haze days, which is closely related to vehicle emissions during the traffic jam. The hygroscopicity of particles was enhanced significantly during the haze days. The probability distribution function of hygroscopic growth factor (GF-PDF) appears a bimodal distribution, with the average GF value of 1.38. The particles number concentration of fine particles in dust days decreased dramatically due to the clear up effect of dust particles. In contrast to the situation during haze events, the hygroscopisity of the particles in dust days displayed a significant decrease, with average GF of 1.17.

31. Airborne In-Situ Measurements of Formaldehyde over California: First Results from the Compact Formaldehyde Fluorescence Experiment (COFFEE) Instrument

Josette E. Marrero, NASA, Moffett Field, CA

Formaldehyde (HCHO) is one of the most abundant oxygenated volatile organic compounds (VOCs) in the atmosphere, playing a role multiple atmospheric processes. Measurements of HCHO can be used to help quantify convective transport, the abundance of VOCs, and ozone production in urban environments. The Compact Formaldehyde FluorescencE Experiment (COFFEE) instrument uses Non-Resonant Laser Induced Fluorescence (NR-LIF) to detect trace concentrations of HCHO as part of the Alpha Jet Atmospheric eXperiment (AJAX) payload. Developed at NASA GSFC, COFFEE is a small, low maintenance instrument with a sensitivity of 100 pptv and a quick response time (1 sec). The COFFEE instrument has been customized to fit in an external wing pod on the Alpha Jet aircraft based at NASA ARC. The instrument can operate over a broad range of altitudes, from boundary layer to lower stratosphere, making it well suited for the Alpha Jet, which can access altitudes from the surface up to 40,000 ft.

We will present results from test flights performed in Fall 2015 over the Central Valley of California. Boundary layer measurements and vertical profiles in the tropospheric column will both be included. This region is of particular interest, due to its elevated levels of HCHO, revealed in satellite images, as well as its high ozone concentrations. In addition to HCHO, the AJAX payload includes measurements of atmospheric ozone, methane, and carbon dioxide. These results will be presented in conjunction with formaldehyde. Targets in the Central Valley consist of an oil field, agricultural areas, and highways, each of which can emit HCHO primarily and generate HCHO through secondary production. Formaldehyde is one of the few urban pollutants that can be measured from space. Plans to compare in-situ COFFEE data with satellite-based HCHO observations such as those from OMI (Aura) and OMPS (SuomiNPP) will also be presented.

32. Evidence for trans-β-IEPOX as the Main Surface-Active Component of Isoprene-Derived SOA Particles

Mary Alice Upshur, Northwestern University, Evanston, IL; and S. T. Martin, R. J. Thomson, and F. M. Geiger

Secondary organic aerosol (SOA) particle formation ranks among the least understood chemical processes in the atmosphere, due in part to a lack of information regarding chemical composition at the gas/aerosol interface. Reactive uptake of surface-active organic oxidation products of biogenic volatile organic compounds (BVOCs) at the gas/aerosol interface can potentially decrease the aerosol surface tension and therefore influence cloud condensation nuclei (CCN) formation. The combination of surface-selective nonlinear vibrational spectroscopy with organic synthesis allows us to gain molecular-level insight into the structure and orientation of terpenes and their oxidation products, enhancing our understanding of SOA formation and surface composition. We utilize a spectroscopic technique called sum frequency generation (SFG) that uses infrared and visible laser light fields, overlapped spatially and temporally at a surface, to probe vibrational transitions within molecules. Use of this technique allows us to analyze the chemical identity of species at the aerosol surface, where interactions with the gas phase can occur. Specifically, we have focused on the synthesis and spectroscopic analysis of the epoxide (IEPOX) and 2-methyltetraol products derived from isoprene oxidation by hydroxyl radicals. Furthermore, dynamic surface tension measurements of synthesized isoprene-derived SOA particle constituents using pendant drop tensiometry demonstrate that these oxidation products depress the surface tension of aqueous droplets, indicating that these compounds may influence the surface chemistry of aerosol particles and have the potential to influence CCN activity.

33. The Role of Organic Species on Aerosol Nucleation and Growth

Jeremiah R. Secrest, Texas A&M University, College Station, TX; and W. Weigang, Y. Zhu, and R. Zhang

New particle formation in the atmosphere is known to affect climate and human health. The fundamental molecular mechanisms of new particle formation and growth are one of the least understood areas in atmospheric chemistry, which hinders regional pollution and global climate change predictions. In this presentation we report experimental results of the effects of organic acids on aerosol nucleation and growth processes. An aerosol nucleation chamber was employed to simulate aerosol nucleation and growth, and the number, size, and chemical composition of freshly nucleated particles as well as the gaseous aerosol precursors were simultaneously measured. Particle number concentrations were measured using a home built Particle Size Magnifier and an Ultrafine Condensation Particle Counter (TSI 3025A). Particle sizes were measured using a nano differential mobility analyzer (TSI 308500), gas concentrations were measured using Ion Drift- Chemical Ionization Mass spectrometry (Balzers QMG 420). Chemical composition of the critical nuclei was measured using Thermal Desorption-Ion Drift-Chemical Ionization Mass Spectrometry. Several different organic acids were employed in our measurements to examine the effects of organic functionality on aerosol nucleation and growth. The atmospheric implications of our experimental results will be discussed.

34. Secondary Aerosol Formation and Growth Analysis Using the mCAGE Environmental Chambers

Manuel A. Salgado, Texas A&M University, College Station, TX; and D. R. Collins

Measurements of secondary aerosol formation and growth rates were made using the mobile Captive Aerosol Growth and Evolution (mCAGE) environmental chambers during the summer of 2015 at a site in the WG Jones State Forest just outside of the Houston, TX metropolitan area. The mCAGE system was developed to monitor and study secondary aerosol production under both ambient and controlled conditions. Initially monodisperse aerosols injected into the chambers are sampled over periods of up to a day as they are altered due to exposure to UV intensity and trace gas concentrations that mimic those just outside. During this study smaller particles (70 nm) were injected and their size tracked to quantify time-dependent growth rate and larger particles (300 nm) were injected to maintain a surface area concentration in the chambers that was comparable to that outside. Initial periods of growth attributed to UV-driven photochemistry during the hours of peak solar intensity were recorded daily. A second distinct period of growth occurred in the hours following sunset. This secondary growth period is thought to be largely due to reaction of biogenic volatile organic compounds (BVOC) such as isoprene and monoterpenes with the nitrate radical (NO3), which is relatively unimportant during the day when it is rapidly photolyzed. Combining nocturnal growth rates with available oxidant and trace gas concentrations provides insight into the controls and significance of the nighttime NO3 chemistry at this site.

35. Assessing the Potential for the Reactions of Epoxides with Amines on Secondary Organic Aerosol Particles

Santino J. Stropoli, Oberlin College, Oberlin, OH; and M. J. Elrod

Nuclear magnetic resonance techniques were used to study the kinetics and products of the reaction of a variety of epoxides with various amines under varying pH conditions. In agreement with a previous finding, the amine-epoxide reactions were found to be water-catalyzed, and not directly dependent on the pH of the reaction environment. At pH values higher than the pKa of the particular amine, the amine-epoxide reactions were extremely efficient, outcompeting hydrolysis reactions even for conditions where water was the solvent and the amine was a relatively low concentration solute. This finding was rationalized by measurements that showed that the relative nucleophilic strength of amines relative to water was on the order of 1000, while the nucleophilic strength of protonated amines (which are predominant when pH < pKa) was negligible. The epoxide carbon substitution environment was found to have a large effect on the measured rate constants (more substituted epoxides had slower rate constants), while the amine carbon substitution environment had a much more limited effect. While the amine-epoxide rate constants are large enough such that amine-epoxide reactions may be kinetically feasible for isoprene-derived epoxides and high amine secondary organic aerosol (SOA) concentrations, most atmospheric amines are expected to be present in protonated form on the largely acidic SOA found in the atmosphere and thus are expected to be largely unreactive towards epoxides.

36. Chlorine-Catalyzed Ozone Loss: UV Absorption Cross Sections, Quantum Yield, and Equilibrium Constant of ClOOCl

David Wilmouth, Harvard University, Cambridge, MA; and J. Klobas and J. G. Anderson

The most important reaction cycle controlling the rate of polar ozone loss is the ClOOCl mechanism, first introduced by Molina and Molina in 1987, in which chlorine peroxide is photolyzed to produce Cl atoms, which destroy stratospheric ozone; the ClO products then self-react to re-form ClOOCl. Importantly, the photolysis reaction is the rate-limiting step in the ClOOCl catalytic sequence, and thus the absorption cross sections and the quantum yield of Cl from ClOOCl photolysis are critical for constraining ozone loss rates in the atmosphere. Significant challenges have been encountered in many past laboratory studies aimed at accurately determining the cross sections of ClOOCl due to difficulty in isolating the spectrum from other absorbing species, either added or produced when chemically forming ClOOCl. In particular, the uncertainty in correcting ClOOCl laboratory spectra for the presence of molecular chlorine, which has an unstructured absorption that peaks at approximately 330 nm, has led to reported ClOOCl cross sections that differed by more than an order of magnitude in some of the most atmospherically relevant wavelength regions. Several recent published studies have served to better constrain the ClOOCl cross section and quantum yield values, but significant uncertainties remain. While the UV cross sections set the photolysis rate, which controls ClOOCl concentrations during the daytime, the equilibrium constant establishes the concentrations of ClOOCl during nighttime. The lab-based, JPL-11 recommended equilibrium constant includes high error bars at atmospherically relevant temperatures (~75% at 200 K) and does not agree well with equilibrium constants empirically determined from in situ atmospheric data. Here we present results of an analysis of available ClOOCl ultraviolet cross section, quantum yield, and equilibrium constant data and present new laboratory spectroscopic results.

37. Aerosols Modeling in the Tropical Atlantic During the Summer of 2009 and Comparisons with Observational Data from the AERosol and Ocean Science Expedition (AEROSE 2009)

Jose M. Tirado, Howard Univ., Washington, DC; and V. R. Morris

Air quality simulations using the chemistry transport model Weather Research and Forecast Chemistry (WRF-CHEM) were performed to study the behavior of aerosol on the tropical Atlantic during the month of July 2009. Aerosols have been show to be at its peak during the month of July in the tropical Atlantic due in part to mineral aerosols (Saharan dust) that travel across the Atlantic to the Caribbean basin and portions of South and North America primarily during the boreal summer. The aim of this project was to observe how dust aerosols affected the tropospheric chemistry of key trace constituents like ozone during its journey to the Caribbean basin. Results from the simulations were compared with trace gases and aerosols data collected on the NOAA ship Ronald H. Brown during the AEROSE 2009 field campaign and data from air quality stations on the island of Puerto Rico. Sensitivity studies were performed to elucidate the contribution of different processes to the chemistry of the area.