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Heterogeneous reactions in the atmosphere have attracted a lot of attention. In particular, reactions involving sea-salt in the form of aerosol droplets, particles, and/or sea-ice have been implicated to significantly affect the chemistry and composition of the marine boundary layer. For example, highly reactive chlorine and bromine atoms resulting from the oxidation of sea-salt halides (Cl^- and Br^-) have been implicated in tropospheric ozone depletion in the arctic and in lower latitude marine regions, as well as the deposition of mercury. While the heterogeneous processing of sea-salt has been studied extensively in laboratory, field and model studies, the mechanistic details behind the release of gas-phase halogens remains unclear and has sparked some interests. Recently there has been attention focused on the interaction of important atmospheric oxidants (e.g., OH and O_3) with halides that reside at the air-particle interface of sea-salt. Such chemical interactions at the surface of particles may lead to unique chemical transformations that can alter current views of known chemical processing of sea-salt particles. There are several laboratory investigations which have investigated the surface reactivity of salts by measuring the reactive loss and/or formation of gas-phase species, indicating that reactions at the interface likely play an important role in aerosol chemistry. The efficacy of such surface-phase chemistry has yet to be elucidated with surface spectroscopy studies. X-ray photoelectron spectroscopy (XPS) is a surface spectroscopy technique with submonolayer resolution. Using XPS, we have investigated changes in the surface chemistry of various alkali halide salts upon exposure to ozone in an ultra-high vacuum (UHV) instrument. Salt samples were either freshly cleaved single crystals which were prepared from a melt, or purified salt crystals/powders pressed into pellets. Upon exposure to ozone, oxygen on the salt surfaces was monitored by measuring the O(1s) photoelectron peak. Initial XPS spectra of salts in the vacuum chamber prior to ozone exposure indicated that the salts were either devoid of oxygen, or contained a small amount of oxygen from strongly adsorbed water under ultra-high vacuum. In all cases, in-vacuo exposure of the salts to ozone lead to an increase in O(1s) photoelectron signal. For NaCl this was surprising since previous studies have been interpreted to indicate that ozone is essentially unreactive towards solid NaCl. The initial uptake of oxygen on the surface gave a broad O(1s) peak, indicating that there are several types of oxygen species on the surface. Reference spectra were obtained for NaClO_2 , NaClO_3 , and NaClO_4 salts. The O(1s) binding energies in these reference spectra increased in series with $\text{NaClO}_2 < \text{NaClO}_3 < \text{NaClO}_4$. Based on these results, it is suggested that the reaction of ozone with pure NaCl initially leads to the formation of NaClO_2 and NaClO_3 , with the formation of NaClO_4 at higher ozone doses. The oxygen uptake upon ozone exposure to alkali bromide and iodide salts were also investigated and compared to reference salts. Future work will incorporate the influence of adsorbed water and subsequent ozone exposures on the surface chemistry of salts.

A11F-0063 0830h POSTER

A Preliminary Field Campaign in the Kathmandu Valley, Nepal: an Urban Photochemistry Study

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The Kathmandu Valley in Nepal experiences severe winter temperature inversions due to its location as a high altitude plateau situated in a semi-enclosed basin. This combined with a rapid increase in population and vehicle ownership has made air pollution of great concern in the valley within the last decade. Only a few sporadic studies have been done characterizing either the meteorology or the chemistry of the valley. This paper presents preliminary surface measurements of O_3 and NO_x ($\text{NO} + \text{NO}_2$) from a field campaign in the Kathmandu Valley during January and February of 2003 and examines the photochemical processes affecting these trace gases. Peak ozone mole fractions ranged from 40 to 100 ppb, while peak NO_x mole fractions were between 20 and 80 ppb. Diurnal variations of ozone, which are substantial, are compared to simultaneous NO_x and micrometeorological measurements to infer factors influencing the production and loss rates

of surface ozone and NO_x . A box model parameterizing the major processes thought to affect valley photochemistry and meteorology is used to further distinguish the relative effect of each precursor emission and model parameter on ozone mole fractions. The magnitude of the simulated maximum or minimum ozone mole fractions appears to be largely dependent on the deposition velocity and ozone levels aloft. The time at which simulated maximum or minimum ozone occurs seems to depend more on ultraviolet dissociation rates and the thickness of the surface and mixed layers.

A11G MCC: 3018 Monday 1020h

A Quarter Century of Satellite Measurements by TOMS II (joint with OS, SA, V)

Presiding: R S Stolarski, NASA
Goddard Space Flight Center; J A
Kaye, NASA Headquarters

A11G-01 1020h INVITED

TOMS and the Identification and Environmental Characterization of Global Dust Sources.

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Although the Total Ozone Mapping Spectrometer (TOMS) was designed for ozone measurements, it has proven to be extremely useful for measuring aerosols. TOMS is especially sensitive to the presence of UV absorbing aerosols such as smoke and mineral dust. An advantage of TOMS as an aerosol sensor is that it can detect aerosols over land as well as ocean surfaces. As a result TOMS has provided us with an unprecedented picture of the global distribution of smoke and dust and their transport paths through the atmosphere. The TOMS absorbing aerosol product has been particularly useful in the study of mineral dust sources which are of great interest because of the important role that dust plays in atmospheric radiation and cloud processes. There were, however, major uncertainties in the sources of dust and the factors affecting dust mobilization and transport. The TOMS product has led to dramatic improvements in our knowledge of the global distribution of dust sources. This, in turn, has enabled us to identify the environmental characteristics of these dusty terrains. TOMS-based studies show that dust sources, regardless of size or strength, can usually be associated with topographical lows located in arid regions with annual rainfall under 200-250 mm. Although the source regions themselves are arid or hyperarid, the action of water is evident from the presence of ephemeral streams, rivers, lakes, and playas. Most major sources have been intermittently flooded through the Quaternary as evidenced by deep alluvial deposits. In this presentation we will review some of these studies and present examples of some of the dust source types that are believed to be characteristic of major sources.

A11G-02 1045h

A long term record of aerosol absorption from TOMS observations

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The discovery of the TOMS capability to detect the presence of absorbing aerosols made possible, for the first time in the history of aerosol sensing from space, the observation of the spatial and temporal variability of carbonaceous and mineral aerosols, over the oceans and the continents. In spite of its qualitative nature, the TOMS aerosol index has been found to be a very useful tool in a variety of applications. In addition to the Aerosol Index product, an inversion procedure was developed to transform the information content of the aerosol index into physically meaningful aerosol parameters: aerosol optical depth and single scattering albedo. Thus, the long term record (1979-1993, 1996-present) of TOMS near UV observations has been used to produce the only global data set on aerosol absorption. The TOMS global climatological record of aerosol absorption optical will be presented and discussed. The results presented here constitute the first long-term nearly global climatological record of aerosol absorption optical depth over both land and water surfaces over regions and times (1979 to present) where ground-based observations are not available.

A11G-03 1100h

TOMS aerosol data and GOCART dust model: Results of a collaborative research

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Satellite data provides the most useful data sets to study aerosol distribution at the global scale over several years. Among the related instruments, the Total Ozone Mapping Spectrometer (TOMS) has the particularity to measure backscattering radiances in the near UV. The advantage of the near UV technique is that aerosol properties can be retrieved over both land and ocean. A difficulty of the technique is the sensitivity of the backscattering radiances to the vertical distribution of absorbing aerosols which is measured continuously at only a few places. Such information has been provided by simulating dust distribution with the Global Ozone Chemistry Aerosol Radiation Transport (GOCART) model. The collaboration GOCART-TOMS went well beyond the delivery of vertical profiles. Some of the key scientific results are: 1) the establishment of the first global inventory of dust source, 2) the characterization of dust emission from the major sources, 3) the study of dust long range transport and dust variability, in particular it was found that dust emission and distribution over Africa is correlated in winter with the North Atlantic Oscillation. Recently, TOMS data has been combined with higher resolution satellite instruments to map exactly all dust sources associated with ephemeral lakes. These results will be presented as well as the methodology developed to obtain them, in particular the use of an equivalent aerosol index which allows direct quantitative comparison with TOMS AI.

A11G-04 1115h

An Expanded UV Irradiance Database from TOMS Including the Effects of Ozone, Clouds, and Aerosol Attenuation

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The TOMS UV irradiance database (1978 to 2003) has been expanded to include 5 new products (noon irradiance at 305, 310, 324, and 380 nm, and noon erythral-weighted irradiance), in addition to the existing erythral daily exposure, that permit direct comparisons with ground-based measurements from spectrometers and broadband instruments. The new data are available on <http://toms.gsfc.nasa.gov>. Comparisons of the TOMS estimated irradiances with ground-based instruments are given along with a review of the sources of known errors, especially the recent improvements in accounting for aerosol attenuation. Trend estimations from the new TOMS irradiances permit the clear separation of changes caused by

ozone and those caused by aerosols and clouds. Systematic differences in cloud cover are shown to be the most important factor in determining regional differences in UV radiation reaching the ground for locations at the same latitude (e.g., the summertime differences between Australia and the US southwest). Regional and seasonal ozone differences are most important at higher latitudes in both hemispheres for causing high values of UVB irradiances.

A11G-05 1130h INVITED

TOMS and Volcanic SO₂: an Important aid to the Understanding of Volcanism and the Atmosphere

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It is impossible to measure gas fluxes from the ground in a major volcanic event, but the TOMS instrument provided the first quantitative measurements of individual stratospheric eruptions, because SO₂ could be measured as well as O₃. The measurements were quickly noticed by scientists, because the masses of sulfur erupted often far exceeded what they expected to find, based on petrology and its supposed constraints, by surprising factors of 10 to 100. This result is still not well understood, and is an important driving idea for volcanologic research. TOMS was applied globally and the explosive volcanic flux of SO₂ to the atmosphere was compiled for the first time using direct measurements – an important input to earth systems analysis. Comparison of TOMS volcanic cloud SO₂ maps with infrared volcanic ash cloud maps showed that there is often spatial separation of gas-rich volcanic clouds emplaced higher in the atmosphere and ash-rich clouds which are lower and which drift in different directions because of windshears. Sequential examination of TOMS data showed that SO₂ masses in volcanic clouds increases for 24 hours or more after eruption. The best explanation of this increase is that ice which forms early in volcanic clouds captures SO₂ which is then released again as the stratospheric ice sublimates. The presentation will document all of the best examples of the discoveries listed above. Volcanologists and those interested in the mitigation of volcanic cloud hazards have repeatedly suggested that geostationary SO₂ and ash sensing capability at higher spatial resolution would provide important new science opportunities. The sensors of the next remote sensing era (MODIS, ASTER, SEVIRI, OMI, ABI) bring us closer to achieving these goals.

URL: <http://skye.gsfc.nasa.gov/index.html>

A11G-06 1150h

Assessing the Ecological Impact of the Antarctic Ozone Hole Using Multisensor Satellite Data

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We have developed a satellite-based numerical simulation for determining the extent to which enhanced solar ultraviolet radiation (UVR) under the springtime Antarctic ozone decrease affects primary production throughout the Southern Ocean. This satellite approach using NASA Sea-viewing Wide Field-of-view Sensor (SeaWiFS) data for chlorophyll and phytoplankton biomass, passive microwave data for sea ice concentration, and Total Ozone Mapping Spectrometer (TOMS) data for total column ozone and cloud reflectivity, circumvents many of the limitations involved with extrapolating point field measurements to larger geographical areas. The satellite data are used to force a physiology-based model of phytoplankton growth in response to UV-B, UV-A, and photosynthetically active radiation (PAR). Comparison with field measurements in the Western Antarctic Peninsula region shows excellent agreement. UVR-induced losses of surface phytoplankton production are substantial, although depth-integrated phytoplankton losses are considerably smaller.

A11G-07 1205h

Evaluation of a Multi-Decadal Simulation of Stratospheric Ozone by Comparison with Total Ozone Mapping Spectrometer (TOMS) Observations

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One key application of atmospheric chemistry and transport models is prediction of the response of ozone and other constituents to various natural and anthropogenic perturbations. These include changes in composition, such as the previous rise and recent decline in emission of man-made chlorofluorocarbons, changes in aerosol loading due to volcanic eruptions, and changes in solar forcing. Comparisons of hindcast model results for the past few decades with observations are a key element of model evaluation and provide a sense of the reliability of model predictions. The 25-year data set from Total Ozone Mapping Spectrometers is a cornerstone of such model evaluation. Here we report evaluation of a three-dimensional multi-decadal simulation of stratospheric composition. Meteorological fields for this off-line calculation are taken from a 50-year simulation of a general circulation model. Model fields are compared with observations from TOMS and also with observations from the Stratospheric Aerosol and Gas Experiment (SAGE), Microwave Limb Sounder (MLS), Cryogenic Limb Array Etalon Spectrometer (CLAES), and the Halogen Occultation Experiment (HALOE). This overall evaluation will emphasize the spatial, seasonal, and interannual variability of the simulation compared with observed atmospheric variability.

A11H MCC: 3016 Monday 1020h

Tropical Cirrus Anvils: Properties and Processes II (*joint with SA, AE*)

Presiding: E Jensen, NASA Ames Research Center; D E Anderson, NASA Headquarters

A11H-01 1020h

The role of cubic ice in dehydration of cold clouds

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Cubic ice is a metastable form of ice with very similar physical properties but different crystal symmetry than normal, hexagonal ice. Below 200 K, water preferentially nucleates to Ic, then transforms to the more stable Ih in minutes to days. As a metastable phase, cubic ice will necessarily have a higher vapor pressure than hexagonal ice. A cloud parcel model shows that nucleation to cubic ice, conversion to hexagonal ice, and the vapor pressure differential together produce larger ice crystals that can more effectively dehydrate air. The modeled effect is largest at 180 to 200 K, just the range most important for the tropical tropopause and polar stratospheric clouds. The cloud model also produces a wider size distribution when cubic ice is included, in agreement with observations. Past observations of cubic ice in the atmosphere are consistent with the modeled behavior.

A11H-02 1035h

Intercomparison of in situ ice water measurements on the WB-57 with radar measurements from the ER-2 during CRYSTAL-FACE

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One of the major goals of the 2002 CRYSTAL-FACE mission was to intercompare and validate measurements of microphysical quantities – ice crystal size distributions, ice mass, and water vapor – by a variety of remote and *in-situ* instruments. However, a direct comparison of remote and *in-situ* measurements is difficult because they involve very different fields of view. CRYSTAL-FACE provided an opportunity for assessing how well such intercomparisons can be made and for determining the type of flight plans that will be necessary for validation of future satellite instruments. During CRYSTAL-FACE, remote and *in-situ* instruments were placed on different aircraft (NASA's ER-2 and WB-57), and the two planes flew in tandem so that the *in-situ* payload flew along the field of view of the remote instruments. We show here, however, that even with this type of careful flight planning, it is not always possible to guarantee with certainty that remote and *in-situ* instruments are viewing the same air parcel. We use ice water data from the *in-situ* Harvard Total Water and Water Vapor instruments, and the remote Goddard Cloud Radar System (CRS), for the flight of July 16, 2002. This date provided the best opportunity for comparison because the planes made multiple passes through cirrus thick enough to be detected by CRS. We show that agreement between Harvard ice water and CRS is a strong function of the horizontal separation and the time delay between the aircraft transects. This type of analysis should guide flight planning for future intercomparison efforts, whether for aircraft-borne or satellite instrumentation.

A11H-03 1050h

Evidence That Most Florida Anvil Crystals Derive From Midtropospheric Aerosols

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