

## A72C-0196 1330h POSTER

**A Chemical Ionisation Mass Spectrometer instrument for the Measurement of Tropospheric HO<sub>2</sub> and RO<sub>2</sub>**

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Laboratory characterisations and field deployments of a Peroxy Radical Chemical Ionisation Mass Spectrometer (PerCIMS) instrument have been performed. During peroxy radical measurements with this instrument, ambient air is sampled through a 20- $\mu$ m diameter orifice into an inlet held at low-pressure, typically 150 torr. Peroxy radicals are converted to other species in the instrument inlet through the addition of NO and SO<sub>2</sub> to the sampled ambient air. The PerCIMS uses this detection technique based on the amplified chemical conversion of ambient peroxy radicals (HO<sub>2</sub> and RO<sub>2</sub>) into a unique ion (HSO<sub>4</sub><sup>-</sup>) following the gas phase reaction of NO and SO<sub>2</sub>. HSO<sub>4</sub><sup>-</sup> ions are then quantified by a quadrupole filter mass spectrometer. Speciated measurements of the total sum of peroxy radicals, HO<sub>2</sub>+RO<sub>2</sub> (HO<sub>x</sub>+RO<sub>x</sub> mode) or the HO<sub>2</sub> component only (HO<sub>2</sub> mode) are achieved through controlling the concentration of the NO and SO<sub>2</sub> added to the inlet. This speciation of peroxy radicals measurement is important in the understanding of the natural oxidation of hydrocarbons that occurs in the troposphere. Both laboratory tests and in situ field measurements during recent aircraft missions and at a radical inter-comparison exercise indicate the PerCIMS to be a feasible field instrument for the fast and accurate evaluation of the concentration of peroxy radicals over a variety of atmospheric conditions.

## A72C-0197 1330h POSTER

**Laboratory Studies to Re-Evaluate the Atmospheric OH Production Rate**

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The major atmospheric source of the hydroxyl radical is the photolysis of O<sub>3</sub> followed by the reaction of O(1D) with H<sub>2</sub>O (1). The major competing loss process for O(1D) radicals is non-reactive quenching with other atmospheric molecules, particularly N<sub>2</sub> (2) and O<sub>2</sub> (3). The rate coefficients for the reactions of O(1D) with H<sub>2</sub>O (k<sub>1</sub>), N<sub>2</sub> (k<sub>2</sub>) and O<sub>2</sub> (k<sub>3</sub>), whose values are critical for calculating atmospheric OH production rates, were measured over a range of atmospherically relevant temperatures to be: k<sub>1</sub>(H<sub>2</sub>O) = (1.49±0.49) 10<sup>-10</sup> exp((80±90)/T) cm<sup>3</sup> molecule<sup>-1</sup> s<sup>-1</sup>; k<sub>2</sub>(N<sub>2</sub>) = (0.195±0.020)exp((125±20)/T); k<sub>3</sub>(O<sub>2</sub>) = (0.365±0.013) exp((20±10)/T). The quoted uncertainties include estimated systematic errors and are at the 95% confidence level. In addition, k<sub>1</sub> was measured relative to k<sub>2</sub> and to k<sub>3</sub> by observing the relative OH concentration produced from reaction (1) with various concentrations of N<sub>2</sub> or O<sub>2</sub> present to be (2.37 ± 0.71) and (1.99 ± 0.91), respectively, at 295 K. Our results indicate that the uncertainties in the calculated atmospheric OH production rate due to the uncertainties in the rate coefficients are less than ± 20%. These results represent a significant improvement in the accuracy and precision of the ability to predict the atmospheric OH production rate.

## A72C-0198 1330h POSTER

**Infrared Spectroscopy and Kinetics of Alkyl Peroxy Radicals**

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The IR spectra of gas phase alkyl peroxy radicals have been measured using an FTIR/long path length absorption apparatus. Radicals are generated by reaction of the parent alkane with atomic chlorine or fluorine followed by association with molecular oxygen. Spectra are confirmed using both experimental and computational techniques. The reaction with nitric oxide is used to titrate the peroxy radical bands attributed to their decrease in intensity strength while new bands stemming from the reaction products are observed. Ab initio methods are used to calculate the band frequencies and intensities. These also assist in assigning the bands to specific vibrational modes of the molecule.

Kinetics of the reactions of alkyl peroxy radicals with NO, NO<sub>2</sub>, and other tropospheric pollutants are measured using a flash photolysis/thermal lensing apparatus in which a xenon flash lamp initiates the reaction mechanism by dissociating molecular chlorine. A pulsed CO<sub>2</sub> laser excites a vibrational band of the peroxy radical. A cw HeNe laser probes the resulting thermal gradient. The strength of the thermal gradient is proportional to the concentration of the absorbing species. The loss of peroxy radical is monitored as a function of time following initiation of the reaction by varying the time between firing of the flash lamp and CO<sub>2</sub> laser. Rate coefficients are measured under pseudo-first order conditions in which the concentration of NO or other reactants are in great excess of the peroxy radical. Spectroscopy results will be presented for ethyl peroxy and larger alkyl peroxy radicals. Kinetic measurements for ethyl peroxy radical will be presented.

## A72C-0199 1330h POSTER

**Measurement of peroxy radicals in the urban atmosphere by PERCA-LIF technique**

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A new instrument has been developed for measuring peroxy radicals (RO<sub>2</sub>) using the Chemical Amplifier-Laser Induced Fluorescence (PERCA-LIF) technique. RO<sub>2</sub> was converted to NO<sub>2</sub> via a chain reaction by the addition of NO and CO in a 1/4" Teflon tube. NO<sub>2</sub> was detected by LIF using Nd:YAG laser (532 nm, 5W at 10kHz). More selective detection of NO<sub>2</sub> is enabled by the LIF than by luminol chemiluminescence because of free from the interference by other oxidants when using luminol. LIF technique can be more sensitive detection of NO<sub>2</sub> than the luminol detector. Optimum conditions were investigated by varying reaction time (i.e. the length of reaction tube) and the concentrations of NO and CO. Maximum chain length of approximately 300 was obtained in dry conditions using a H<sub>2</sub>O/O<sub>2</sub> simultaneous photolysis method. Experiments were performed to characterize the dependence of the chain length on humidity for this instrument. In August 2002, RO<sub>2</sub> measurements were performed in Osaka using this method. Maximum concentrations of RO<sub>2</sub> in the daytime were approximately 100 pptv. Nighttime observations were also conducted and significant concentrations of RO<sub>2</sub> were detected just after the sunset. Existence of formation processes in the dark condition was investigated.

## A72C-0200 1330h POSTER

**Kinetics of the Reaction of OH with Peroxynitric Acid (HO<sub>2</sub>NO<sub>2</sub>) between 218 and 358 K**

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Peroxynitric acid (PNA), HO<sub>2</sub>NO<sub>2</sub>, is formed in the atmosphere by the HO<sub>2</sub> + NO<sub>2</sub> + M reaction. Removal of HO<sub>2</sub>NO<sub>2</sub> in the upper troposphere and lower stratosphere occurs mainly via photo-dissociation and reaction with OH radicals. We present our results on the kinetics of OH radicals with PNA in the gas-phase as a function of temperature (T=218-358 K) obtained by using pulsed laser photolysis (at 193 and 248 nm) to generate OH and laser-induced fluorescence for monitoring its temporal profiles. The concentration of PNA in the reactor was measured by UV absorption. The unavoidable impurities present in the PNA samples (H<sub>2</sub>O<sub>2</sub>, HNO<sub>3</sub> and NO<sub>2</sub>) were quantified by UV absorption or mass spectrometry to account for the contribution of the reactions of OH with these impurities to the measured rate coefficients. The bimolecular rate coefficients for the OH + PNA reaction was found to be independent of pressure between 10 and 125 Torr of Helium. The temperature dependence of the rate coefficients for the OH + PNA reaction showed non-Arrhenius behavior over the range studied. The discrepancy between our rate coefficients and those previously reported in the literature will be discussed. The implication of our measured rate coefficients to the upper troposphere and lower stratosphere will be also discussed.

## A72D MCC: 125 Sunday 1330h

**Advances in Physics and Chemistry of Clouds I (joint with H, GC)**

**Presiding:** G Kramm, University of Alaska, Fairbanks; R M Rasmussen, National Center for Atmospheric Research

## A72D-01 1335h INVITED

**Turbulence-Particle Interactions in Clouds: Theory, Measurements, and Implications**

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Droplets in a cloud often are not spatially distributed in a perfectly random manner: instead, they are clustered on various scales. However, most theories of cloud processes, such as collision-coalescence and radiative transfer, are based on the assumption that droplet positions are uncorrelated. Correlations in droplet positions can be caused by turbulent mixing of cloudy and clear air, by the inertial response of cloud droplets to fluid accelerations (preferential concentration), or even by gravitational sedimentation in still air. The degree of spatial correlation, quantified by the pair correlation function, is scale dependent and is a function of such variables as droplet size, energy dissipation rate, and turbulence Reynolds number. Spatial correlations measured in turbulent clouds with a Fast FSSP are observed to be strongest at centimeter scales and below and the pronounced clustering on these scales is consistent with the inertial clustering hypothesis.

Clustering on sub-centimeter scales is especially relevant to processes that occur on those scales, the collision-coalescence process being archetypal. For example, when droplets are not distributed with perfect randomness the coagulation equation (stochastic collection equation) used in cloud physics will give erroneous results. The coagulation equation can be modified, however, if the droplet pair correlation function is known. It follows that if the pair correlation function can be determined theoretically or measured directly the modified collision rate can be properly accounted for. As a specific example, an analytical form of the modified coagulation equation appropriate for a turbulent mixing zone, based on the Broadwell-Breidenthal mixing model, can be formulated and leads to enhanced droplet collision rates.

## A72D-02 1355h

### Cloud properties inferred from surface air temperature changes associated with the presence of ground-based cloud types over land areas

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Surface air temperature changes associated with the presence of clouds (TC), defined as the difference between surface air temperature with and without clouds, are studied using the long-term ground-based cloud datasets from the U.S. and former USSR national meteorological station networks. An approach is developed to separate the cloud longwave effect-related surface air temperature change (LWTC) and the cloud shortwave-effect related surface air temperature change (SWTC) from TC specific for each cloud type. The magnitudes of LWTC and SWTC is then analyzed for each commonly occurring cloud type in the context of their relationships with cloud radiative properties, including cloud amount, albedo and emittance as well as with two other internal climate variables affecting these relationships: near-surface air humidity and snow cover extent. We hope that the cloud type radiative properties and the cloud interaction within the climatic system explored in this empirical study can help better understand the role of clouds in contemporary climatic change. This approach is based mostly on the surface cloud observing system, which is fundamentally different from the satellite observing system.

## A72D-03 1410h INVITED

### Active Remote Sensing of Clouds

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A variety of research tools are available for studying the macrophysical and microphysical properties of clouds, information of crucial importance to modeling their effects on the radiation balance of the earth/atmospheric system. In this report we summarize the advantages of active ground-based, and planned satellite-borne, remote sensing systems. Although a variety of microwave radar and laser-radar (lidar) techniques are available, it is through the synergism gained from coordinated active and passive remote sensors that the greatest knowledge is gained. The history of this multiple remote sensor approach will be reviewed, and the latest results highlighted.

## A72D-04 1430h

### Measurements of Ice Water Content in Tropopause Arctic Cirrus during SOLVE (SAGE III Ozone Loss and Validation Experiment)

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The greatest uncertainty in the prediction of future global climate arises from our incomplete knowledge of the interaction of clouds with solar and terrestrial radiation. Cirrus clouds are of special interest due to their location in the upper troposphere, where their unique optical and microphysical properties contribute to local radiative heating, as well as creating a surface for heterogeneous chemical reactions. In this paper, we present observations of high-latitude cirrus clouds which were made during the SOLVE (SAGE III Ozone Loss and Validation Experiment) mission based in Kiruna, Sweden throughout the winter and spring of 1999-2000. The data used in this study were obtained with three instruments: an open-path tunable diode laser hygrometer, a FSSP-300 Particle Measurement System, and a closed-path tunable diode laser hygrometer, which sampled particulate water. The water

content of each cloud encounter was calculated from size distributions obtained by the FSSP. As expected, the structure of the FSSP observations and the closed-path hygrometer particulate water data were similar. However, the closed-path tunable diode laser hygrometer typically reported considerably more condensed water than that calculated from the FSSP particle data. The excess particle water seen by the laser hygrometer provides evidence for larger cirrus cloud ice crystals outside the range of the FSSP (i.e. greater than 20  $\mu$ m in diameter).

## A72D-05 1445h INVITED

### A Step Towards an Advanced Parameterization of Cloud Microphysical Processes

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Consideration of cloud microphysical properties and processes in atmospheric models usually requires reliable and accurate parameterizations. For describing all hydrometeor types by size distribution functions and corresponding budget equations comprising a multitude of processes in an adapted manner is by far too costly.

An alternative is to only deal with certain integrals (i.e. moments of the size spectra as, e.g., water contents) and their tendency equations. Moreover, the parameter formulae should comply with the natural situation of having smaller (cloud) and larger (precipitation) particles which interact by collisions in a complex way.

Many years ago this idea has been elaborated by Kessler (1969) for liquid (warm) clouds. Kessler presented a rate equation for the transformation of cloud water content to rainwater mass (autoconversion) which relies on high intuition and another one for accretion, i.e. for the increase of rainwater content by mutual collection of cloud droplets by raindrops, which is based on a simplistic evaluation of the collection integrals of the spectral budget equation for drops. This first approach to parameterize the evolution of rain water from cloud water is a very important one since almost all clouds start as liquid clouds.

For a long time and also to date these so-called Kessler formulae were the only parameterization available for warm cloud processes. In adopting this idea corresponding formulations have also been derived and extensively applied for mixed and ice cloud microphysics.

The drawback of Kesslers formulation is that it only uses (cloud and rain) water contents such that a differentiation between continental and maritime clouds exhibiting very different size spectra but identical water contents is not possible.

To overcome this deficiency and to include typical cloud characteristics several authors extended Kessler's idea by formulating - in addition to the rates of change of mass contents - rates for the corresponding number densities. As a closure condition representative size distributions (e.g. Marshall Palmer) have then to be considered. Unfortunately this procedure has mostly not uniformly been performed for all hydrometeor types and processes and critical parameters have been set constant - an assumption which is crucial.

Surprisingly these suggestions were disregarded in case of the warm rain processes autoconversion and accretion which are basic in the development of each (warm) cloud. Some years ago these mechanisms, however, have been formulated on the basis of the stochastic collection equation resulting in spectral and integral rate equations. Moreover, by reasonable approximations advanced rate equations for the number and mass densities changing by autoconversion and accretion have been developed taking into account different cloud spectrum characteristics.

In that way a complete and consistent set of equations for the time rate of change of number as well as mass densities of cloud and rain, snow and ice particles can be formulated covering the whole range of processes occurring in warm, mixed and ice clouds.

The presentation addresses all items mentioned and concludes with results of numerical experiments which demonstrate the difference between usual and the advanced parameterizations in case of convective clouds.

## A72E MCC: 102 Sunday 1330h

### Transport and Effects of Anthropogenic Pollutants: ACE-Asia I (joint with OS, GC)

Presiding: T Bates, NOAA Pacific Marine Environmental Laboratory; B Huebert, University of Hawaii; P Russell, NASA Ames Research Center

## A72E-01 1340h

### Aerosol In Asian Outflow: Correspondence of Size-dependent Aerosol Volatility to Chemistry, Humidity Growth, and Absorption

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During Spring 2001 we participated in the ACE-Asia project, studying the properties of Asian aerosol advected over the North Pacific from the NCAR C-130 aircraft. We also had a smaller suite of instruments aboard the NASA P-3 during TRACE-P. These sequential experiments with similar instrumentation provided a unique opportunity to investigate outflow from East Asia between 5°N and 50°N during the same season. Size distributions were obtained using a variety of instruments, including radial differential mobility analyzer (RDMA), a laser optical particle counter (OPC) and an aerodynamic particle sizer (APS). The RDMA and OPC were operated with heating of the inlet stream, cycling between unheated, 150°C, and 300°C in order to infer size dependent volatility and refractory properties. Size-resolved chemical composition was provided by cascade impactors and single-particle electron microscopy. Bulk ionic composition of 0.1 to 1.5  $\mu$ m particles was measured rapidly (5 min) with a Particle Into Liquid Sampler (PILS). Submicron and total aerosol optical properties including scattering, absorption, and humidity dependence [ $f(\text{RH})$ ] were measured with a suite of 4 nephelometers and 2 Particle Soot Absorption Photometers (PSAP).

While the thermal treatment cannot directly differentiate between dust and elemental carbon, we found that the submicron tail of the dust distribution was consistently enough shaped that we could isolate the refractory components of the accumulation mode aerosol from the dust. That enables us to calculate the contributions of each to optical extinction and single scatter albedo.

Given the chemical composition of the aerosol, we can calculate the refractive index, which we use to correct the OPC calibration. After this correction, we found that the volatile volume corresponded closely to measured sulfate and nitrate composition. Refractory volume in the accumulation mode (variable, but generally <0.5 $\mu$ m) was well correlated with both PSAP absorption and EC measurements, though it appears that only 30 to 50% of the refractory volume is carbon.

Size-dependent chemical composition was also used to approximate  $g(\text{RH})$ , particle growth as a result of water condensation as humidity rises. Mie scattering calculations gave good agreement with measured  $f(\text{RH})$ . In general,  $f(\text{RH})$  behavior simply mirrored the relative contributions of dust (non-hygroscopic) and accumulation mode (hygroscopic) surface areas. However, variations in accumulation mode composition did affect  $f(\text{RH})$ . Those changes were reflected in the volatility of accumulation mode particles.