

whether this trend is a result of natural variability or is related to greenhouse warming, a mechanistic model is developed that provides a simple analogue of the NAM. The model represents the interaction between the zonally symmetric flow and planetary waves for two meridional modes of variation. Greenhouse warming is specified in the model by varying the radiative equilibrium meridional temperature gradient. Variations in the second meridional mode qualitatively resemble those of the observed NAM. Model runs that correspond to larger greenhouse gas concentrations yield on average higher NAM index values. Additional model runs have been performed to investigate the dependence of NAM variability on the height of the upper boundary. These simulations reveal that the amplitude of the NAM signal gradually weakens as the upper boundary is lowered below 30 km. With the upper boundary at 10 km, essentially no NAM variability exists in the model. These results from the mechanistic model will be discussed in relation to other more complex Mechanistic Circulation Model (MCM) and General Circulation Model (GCM) simulations.

A31A-12 1145h

Relationship Between Climate and Atmospheric Chemical Species Modulated by Arctic Oscillation

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The relationship between the interannual variability of climate variables, such as geopotential height, temperature and zonal wind, and the interannual variability of chemical species, such as ozone, methane and nitrous dioxide, in the northern winter high latitudes was investigated using two 15-year equilibrium simulations of the University of Illinois at Urbana-Champaign Coupled Climate/Chemistry Model and the NCEP/CPC reanalysis data. Temporal Empirical Orthogonal Function analysis was used to define the dominant (first) mode of the different quantities interannual variability. A sensitivity of the first mode to the solar UV radiation enhancement in at the top of the atmosphere was also studied. Analysis showed that in the polar winter, when the Arctic Oscillation is the dominant pattern of the geopotential height variability, the relationship between the different quantities first modes could be recognized in correspondence with a geopotential height phase change. Namely, in the stratosphere a change in the geopotential height at one height occurs with: (1) changes of the same sign in temperature and ozone at lower altitudes, and a temperature change of opposite sign at the higher altitudes; (2) the same sign changes in methane and nitrous oxide at the same height, and the opposite methane and nitrous oxide changes at lower altitudes. The imposed enhancement of the solar UV radiation, which affects the daytime photodissociation rates of the chemical reactions and the radiative heating of the atmosphere, did not modify much the relationship between variables. However, the correlation between the dominant modes in the middle stratosphere increased for most variables and an appearance of the years with intensive polar vortex decreased. Also, the polar middle and lower stratosphere become slightly cooler, and the concentrations of the ozone and methane decreased there. The amplitude of the variables interannual variability decreased and the polar circulation becomes more zonal.

A31B MC: 133 Wednesday 0830h

Advances in Aerosol Science and Technology I

Presiding: S S Cliff, University of California; R Arimoto, New Mexico State University

A31B-01 0830h INVITED

Three Compact, Robust Chemical Characterization Systems Suited To Sensitive, High Time Resolution Measurements Of Atmospheric Aerosols

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In the past decade, the advancement of compact, robust and sensitive instrumentation to measure the chemical characteristics of atmospheric aerosols has lagged behind their physical characterization. There is a need for chemical instrumentation with these three qualities for use on airborne platforms and at infrequently attended ground level surveillance sites. Now chemical techniques are appearing that promise to fill this need.

We discuss three chemical characterization systems that are emerging in atmospheric chemistry and climate research applications. These are: (i) the Aerodyne mass spectrometer for real time measurement of particle composition and two post-collection analysis techniques (ii) non-destructive, multi-elemental chemical analysis of size-resolved samples by high spatial resolution synchrotron x-ray and proton beams (S-XRF/PIXE/PESA/STIM) (iii) single particle characterization by automated scanning electron microscopy with energy-dispersed detection of X-rays (SEM/EDX).

The key to post-collection analysis is automated aerosol sizing and collection systems and automated chemical analysis systems. Together these techniques provide unique, comprehensive information on the organic and inorganic composition and morphology of particles and yet are easy to deploy in the field. The sensitivity of each technique is high enough to permit the rapid sampling needed to resolve spatial gradients in composition from a moving platform like the Battelle Gulfstream-159 aircraft, traveling at 100m/s.

A31B-02 0850h INVITED

Mass Independent Isotopic Compositions of Aerosol Sulfate and Nitrates

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For nearly a half-century stable isotope ratio measurements have been utilized as a tool to understand sources, fates, and transformation mechanisms of atmospheric molecules. Carbon and oxygen ($\delta^{13}\text{C}$ and $\delta^{18}\text{O}$) measurements of CO_2 have been instrumental in providing specific details of the carbon cycle. Without these measurements, understanding of the carbon cycle and transfer rates between reservoirs would be considerably diminished. Deuterium and oxygen isotopic measurements of atmospheric water has similarly enhanced the ability to model the atmospheric and geochemical recycling of the hydrologic cycle. Other molecules investigated include, for example, CO , CH_4 , N_2O , SO_4 , NH_3 , and Cl . The ability to interpret these high precision isotope ratio measurements relies upon a fundamental understanding of the basic physical-chemical processes which produce the alteration of the stable isotope ratio. Such processes typically include thermodynamics (viz a viz isotope exchange), kinetics, and evaporation-condensation. Though the mechanism by which these alterations occur, they all depend in some fashion upon mass differences in the isotopically substituted atoms.

In 1983, Thiemens and Heidenreich (1) demonstrated that a chemical process is capable of producing an alteration of stable isotopes which was independent of mass. Subsequent to that time, it has been shown that measurements of mass independent isotopic compositions provide a new view of atmospheric process which may not be derived from single isotope ratio measurements (reviews by (2), (3)). In the past few years, mass independent isotopic compositions have been utilized to understand ancient atmospheres on both Earth and Mars (review by (4)).

It has been known for decades that atmospheric sulfate is an extraordinary species. It participates in climate change in its capacity as a cloud condensation nuclei and it is a human and environmental health hazard. By the same token, aerosol nitrate is an environmental health hazard and also is known to produce changes in biodiversity. In spite of decades of high quality measurements and models there remain significant gaps in understanding. For example, the relative proportion of homo vs heterogeneous SO_2 oxidation to sulfate and the extent of long range transport are issues where further insight is needed. Likewise, these are key issues for nitrate.

For both nitrate and sulfate, mass independent isotopic compositions have recently been observed. These anomalous isotopic signatures have now provided a new measure to widen our understanding of the atmospheric sulfur and nitrate cycles. The mechanisms of chemical transformation and long range transport have been further elucidated. In addition, measurement of these species in polar ice samples have provided a means by which global oxidative processes have varied on 100,000 year time scales.

(1) Thiemens and Heidenreich (1983) Science 219, 1073.

(2) Thiemens (1999), Science 283, 341.
(3) Weston (1999) chem. Rev. 99, 2115.
(4) Thiemens, Savarino, Farquhar, Bao (2001) 34, 645.

A31B-03 0910h INVITED

The new MODIS -Terra, and the proposed COBRA mission: First global aerosol distribution and properties over land and ocean, and plans to measure global Black Carbon absorption over the ocean glint

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The MODIS instrument was launched on the NASA Terra satellite in Dec. 1999. Since last Oct, the sensor and the aerosol algorithm reached maturity and provide global daily retrievals of aerosol optical thickness and properties. MODIS has 36 spectral channels in the visible to IR with resolution down to 250 m. This allows accurate cloud screening and multi-spectral aerosol retrievals. We derive the aerosol optical thickness over the ocean and most of the land areas, distinguishing between fine (mainly man-made) and coarse (mainly natural) aerosol particles. New methods to derive the aerosol absorption of sunlight are also being developed. These measurements are used to track different aerosol types, sources, transport and the radiative forcing at the top and bottom of the atmosphere. However MODIS or any present satellite sensor cannot measure absorption by black carbon over the oceans, a critical component in studying climate change and human health. For this purpose we have proposed a satellite mission that observes the ocean sun-glint. Using both on glint and off glint measurements of spectrally polarized light, for wavelengths up to 2.1 μm , we can derive the aerosol absorption.

A31B-04 0925h

Comprehensive Laboratory Measurements of the Emissions From Fires in African and Other Globally Significant Fuels Measured by FTIR, PTR-MS, and GC

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Biomass burning is one of the most important influences on the global atmosphere. Field experiments have yielded much useful knowledge about fires, but are often limited by lower S/N, lack of fuels data, and the challenges imposed by operating in remote tropical locations. In large-scale laboratory experiments at the U. S. Forest Service (USFS) Fire Sciences Laboratory we captured and measured all the emissions produced by 54 separate fires in 16 fuel types from southern Africa, Indonesia, Canada, the U. S., and Germany. Fuels included Dambo grass, Miombo litter, and Indonesian rice straw and peat. The fires were carefully simulated to match (as closely as possible) actual fires observed primarily during SAFARI-2000 and in Indonesia. Fuel C:H:N content was measured and fuel mass loss was continuously monitored. Total pressure, temperature, and flow of trace gases was monitored at the sampling platform in the stack above the fires. Trace gases were speciated by an impressive array of instrumentation. Both a closed cell and open-path FTIR were deployed by the UM group to quantify CO_2 , CO , CH_4 , NMHCs, oxygenated VOCs, NO_x , HCN , and NH_3 above ppb levels yielding a broad overview of the major smoke constituents. A proton-transfer reaction mass spectrometer (PTR-MS) from MPI was used to measure VOCs at

ppt levels. NDIR instruments independently measured CO₂ and CO. Canister sampling with GC analysis by MPI, USFS, and UC Irvine also measured CO₂ and CO as well as hydrocarbons and halogenated hydrocarbons. Particles were sampled on quartz and Teflon filters to measure the emission factors for PM_{2.5} and elemental and organic carbon. These results constitute the most comprehensive measurements of fire emissions to date and also the first intercomparison between FTIR and PTR-MS. PTR-MS can quantify the total VOC (with proton affinity higher than water) present at each mass up to 200 a.m.u. at ppt levels. At ppb levels most molecules have multiple IR peaks so FTIR is ideally suited for compound identification. The combination of these two techniques is very powerful. Results include confirmation of the high emissions of oxygenated organic compounds and the relatively low emissions of ammonia by African fires suggested by our airborne FTIR measurements during SAFARI-2000. In addition, numerous compounds were quantified that were below our detection limit in the field campaign thus providing a more complete understanding of these important types of biomass fires.

A31B-05 0940h

Single Particle Source Profiles of Gasoline and Diesel Powered Vehicles, Biomass Burning and Coal Combustion Exhaust Emissions

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Vehicular exhaust, biomass burning, and coal combustion are three significant aerosol sources that have local to global impacts on the earth's atmosphere. They may also contribute to health effects as they can emit carcinogenic species such as polycyclic aromatic hydrocarbons (PAH) and trace metals including beryllium and vanadium. In these source characterization studies, combustion products were diluted to near ambient temperature and pressure using a two stage dilution source sampler. Diluted exhaust emissions were analyzed with an aerosol time-of-flight mass spectrometer (ATOFMS) obtaining real-time measurements of single particle size and chemical composition. In addition, samples were collected using a micro-orifice uniform deposit impactor (MOUDI), which was operated in a manner compatible with advanced chemical analysis techniques, for size segregated mass concentrations. Due to the importance of these particle sources to the atmosphere, differentiating these emissions from each other and other particle sources is essential. Since ATOFMS is a relatively new single particle analysis technique, source characterization experiments are needed to determine qualitative signatures of specific particulate sources for their ambient identification. ATOFMS single particle mass spectra will be discussed introducing chemically distinct single particle types emitted from these combustion sources. Numerous particle types are emitted from each source, as indicated by distinct chemical associations on the single particle level. Examples include, the chemical associations of vanadium with organic carbon (OC) in gasoline powered vehicle emissions, calcium with black carbon (BC) in diesel powered vehicle emissions, beryllium and boron with BC in coal combustion emissions, and potassium with OC from biomass burning emissions. Most importantly, the overall particle type distributions from each source differ significantly. Finally, complementary MOUDI mass distribution data will be used to determine the relative fractions of these particle types to the overall particulate mass emissions from these tests. These results will be presented in terms of single particle source profiles for these environmentally important combustion aerosol sources.

A31B-06 0955h

Analysis of Aerosols Collected in Kyoto, Japan and Cheju, Korea in Spring 2001

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Investigation into the composition of aerosol particles is essential to comprehend the effects they have on both our climate and our health. Laser desorption/ionization time of flight mass spectrometry has been used to inspect both the organic and inorganic composition of aerosol samples collected in Kyoto, Japan and Cheju, Korea. Samples were collected in eight size fractions on an aluminum substrate during the ACE-Asia sampling event Spring 2001. The size fractions collected are inlet to 5, 5 to 2.5, 2.5 to 1.15, 1.15 to 0.75, 0.75 to 0.56, 0.56 to 0.34, 0.34 to 0.24, and 0.24 to 0.09 microns. In addition to being segregated by size, these samples were also collected as a function of time to aid in the determination of the source of the particulate matter. Analysis by LDI-TOFMS in negative ion mode shows differences in the inorganic composition, specifically in nitrates and sulfates, between aerosol sizes. Variations in the organic component of these aerosols, specifically polycyclic aromatic hydrocarbons (PAHs), are examined in positive ion mode. The impactor/LDI-TOF-MS arrangement provides a method for time and size resolved fingerprinting of the collected aerosols.

A31B-07 1030h INVITED

Mapping Organic Composition in Individual Atmospheric Particles

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The organic composition of aerosols can play an important role in determining the amount of water taken up by particles, the fraction of light absorbed in particles, and the heterogeneous reactions catalyzed on particles. While identifying individual organic compounds is impractical for many atmospheric applications, the characterization of organic functional groups provides critical information about compound polarity and solubility. Sampling organic particles from aircraft requires innovative techniques to collect sufficient sample mass for analysis.

The technique proposed here requires highly sensitive detection of a small number of organic molecules in order to identify functional groups in individual aerosol particles. Particles are collected by impaction on a thin film, using a 15-minute sampling period. The impacted particles are analyzed at atmospheric pressure with soft X-ray spectroscopy at the Advanced Light Source to characterize the presence of different functional groups on individual particles as small as 0.1 micron diameter. Spatial resolution of the spectroscopic images also allows mapping of heterogeneities within the particle.

Particle samples taken aboard the NCAR C130 research aircraft near Japan during ACE-Asia and near St. Croix during PELTI illustrate important differences between the relative amounts of carbonyls, aromatics, and other organic compounds present in particles in clean and polluted air masses. Spatial resolution of 0.1 micron allows us to map the particle organic composition, revealing complex inorganic and organic internal mixtures and heterogeneous structures.

A31B-08 1045h INVITED

Advanced Elemental and Isotopic Characterization of Atmospheric Aerosols

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Recent sampling and analytical developments advanced by the project team enable the detailed elemental and isotopic fingerprinting of extremely small masses of atmospheric aerosols. Historically, this type of characterization was rarely achieved due to limitations in analytical sensitivity and a lack of awareness concerning the potential for contamination. However, with the introduction of 3rd and 4th generation ICP-MS instrumentation and the application of state-of-the-art "clean-techniques", quantitative analysis of over 40 elements in sub-milligram samples can be realized. When coupled with an efficient and validated solubilization method, ICP-MS approaches provide distinct advantages in comparison with traditional methods; greatly enhanced detection limits, improved accuracy, and isotope resolution capability, to name a few. Importantly, the ICP-MS approach can readily be integrated with techniques which enable phase differentiation and chemical speciation information to be acquired. For example, selective chemical leaching can provide data on the association of metals with major phase-components, and oxidation state of certain metals. Critical information on metal-ligand stability can be obtained when electrochemical techniques, such as adsorptive cathodic stripping voltammetry (ACSV), are applied to these same extracts.

Our research group is applying these techniques in a broad range of research projects to better understand the sources and distribution of trace metals in particulate matter in the atmosphere. Using examples from our research, including recent Pb and Sr isotope ratio work on Asian aerosols, we will illustrate the capabilities and applications of these new methods.

A31B-09 1100h

The use of STIM and PESA to respectively measure profiles of aerosol mass and hydrogen content across Mylar rotating drum impactor samples

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A method has been developed for measuring profiles of aerosol mass on thin (480 mg/cm²) Apiezon-L coated Mylar films employed in rotating drum aerosol impactor samplers using the ion beam analysis technique Scanning Transmission Ion Microscopy (STIM). The greased Mylar films are excellent impaction substrates and possess excellent uniformity in projected density making them an ideal substrate for STIM analysis. The uniformity in projected density of a film enables STIM with a 3 MeV proton beam to produce profiles of aerosol mass with an accuracy of better than 90% and a mass sensitivity approaching 10 mg/cm². Further, we have extended Proton Elastic Scattering Analysis (PESA) to the same films, achieving measurement of an organic-surrogate. Although the films contain 20 mg/cm² hydrogen, the spatial uniformity in film hydrogen content enables PESA with a 3 MeV proton beam to produce profiles of hydrogen arising solely from the aerosols with an accuracy to within ± 1 mg/cm² and a mass sensitivity of 1 mg/cm². These measurements, when combined with synchrotron-x-ray fluorescence (S-XRF) measurements on the same film allow mass closure, sum of species versus measured mass - a key quality assurance protocol, to be approached. All three techniques were applied to very fine and ultra-fine particles collected in Fresno, CA, November, 2000 by slotted DRUM samplers. Temporal resolution in the resulting profiles was * 6 hours. The dramatic changes in composition versus size and time, and new types of elemental correlations unseen in PM_{2.5} filters, will be major assets in correlating aerosols and health impacts, visibility degradation, and the effects of aerosols on climate.

A31B-10 1115h

Airborne Sunphotometry in Support of the Chesapeake Lighthouse and Aircraft Measurements for Satellites (CLAMS) Experiment, 2001

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As part of the Chesapeake Lighthouse and Aircraft Measurements for Satellites (CLAMS) experiment, July 10 - August 2, 2001, the 14-channel NASA Ames Airborne Tracking Sunphotometer (AATS-14) was operated successfully aboard the University of Washington CV-580 during 10 research flights, totaling 45.09 flight hours. The CLAMS campaign was a clear sky, short-wave (SW) closure campaign sponsored by CERES, MISR, MODIS-Atmospheres, and the NASA/GEWEX Global Aerosol Climatology Project (GACP), and entailed measurements from the Chesapeake Lighthouse research platform, several land sites, 6 research aircraft and the TERRA satellite. Among the CLAMS research goals were the validation of satellite-based retrievals of aerosol properties, vertical profiles of radiative fluxes, temperature and water vapor.

AATS-14 measures the direct solar beam transmission at 14 discrete wavelengths (354-1558 nm), yielding aerosol optical depth (AOD) spectra, columnar water vapor and columnar ozone. Differentiation of AOD (CWV) with respect to altitude in favorable flight patterns, allows the derivation of aerosol extinction (water vapor density). During coordinated flights of the UW CV-580, AATS-14 measured full column aerosol optical depth spectra at exact TERRA overpass time on at least 7 occasions. For five of these opportunities, AOD at 499nm was at or below 0.1. During TERRA overpass time on July 17, 2001, AATS-14 measured the highest AOD encountered during the entire experiment (ca. 0.48 at 499nm), including a horizontal gradient in AOD of more than 0.1 over a horizontal distance of ca. 80 kilometers.

In this paper, we will show first sunphotometer-derived results regarding the spatial variation of AOD and CWV during TERRA overpass time at key locations for the CLAMS experiment. Preliminary comparison studies between our AOD/aerosol extinction data and results from (i) extinction products derived using in situ measurements aboard the UW CV-580 and (ii) AOD retrievals using the Multi-angle Imaging Spectro-Radiometer (MISR) aboard the TERRA satellite will also be presented.

A31B-11 1130h

Measurements of Absorbing Aerosols Using In Situ and Remote Sensing Techniques

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Reliable measurements of light absorption by aerosol particles are essential for an accurate assessment of the climate radiative forcing by aerosol particles. Depending on the absorption properties, the radiative forcing of the aerosols may change from a cooling to a heating effect. New techniques for the remote sensing of aerosol absorption over land and ocean are developed and applied in combination with in situ measurements for validation and addition of complementary information. Spectral measurements show the effects of aerosols on absorption of light from the UV to the near infrared. Depending on particle size and structure, there is a significant absorption component that must be accounted for the radiative forcing in the near infrared. Remote sensing results from MODIS and from the CLAMS field experiment, as well as in situ validation data will be discussed.

A31B-12 1145h

Global Modeling of Tropospheric Aerosols by LLNL IMPACT and Comparisons with Field Measurements*

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A new version of LLNL IMPACT (Integrated Massively Parallel Atmospheric Chemical Transport) model driven by NCAR MACCM3 has been used to study the global aerosol cycle in the troposphere. This new version with compact chemical mechanisms (~21 prognostic species) is aiming at developing the aerosol climatology and exploring the impact of aerosols on climate variability and climate change. We will present global distributions of four major aerosol components (sulfate, carbonaceous, dust and sea salt) and their seasonal variations. By applying the monthly averages of OH, HO₂, and H₂O₂ from previous IMPACT simulations with full chemistry (~100 prognostic species), sulfate is formed through both gas and aqueous oxidation from SO₂ and DMS emissions. Other aerosol types are assumed to be injected into the global model in the particulate form. Individual aerosol concentrations are compared with field measurements at different geographical locations to validate the accuracy of the model.

*Work performed under the auspices of the U.S. Department of Energy by the University of California, Lawrence Livermore National Laboratory under contract No. W-7405-Eng-48.

A32A MC: Hall D Wednesday 1330h

The Arctic Oscillation and the North Atlantic Oscillation: Mechanisms, Coupling, and Climate Change (*joint with OS*)

Presiding: T Furevik, Nansen

Environmental and Remote Sensing Center

A32A-0023 1330h POSTER

Does the NAO index represent zonal circulation? The influence of the NAO on North Atlantic surface temperature

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We investigate the influence of zonal and meridional flow on surface temperature in the North Atlantic/European region. The degree to which the North Atlantic Oscillation (NAO) index reflects these two different flow types is considered, as is the relationship between the NAO index and surface temperature. Zonal

and meridional circulation indices extending back to the early 19th or 18th centuries are based on surface pressure observations from the North Atlantic and Europe and on an Empirical Orthogonal Function (EOF) analysis of European surface pressure from 1845-1995. The NAO index appears to integrate aspects of both zonal and meridional flow types. The surface temperature changes associated with the NAO index is composed of a quadrupole correlation pattern, showing positive correlations over Europe and the Sargasso Sea and negative correlations over north-west Africa and the Greenland/Labrador Sea region. It appears that the relationship between the NAO index and temperatures downstream of the Atlantic is associated with zonal flow, whereas the influence of the NAO on temperatures upstream is more closely linked to meridional flow patterns. Running correlations indicate that while there is no obvious link between the NAO index and the secular temperature trend, the second principal component of temperature is closely linked to atmospheric circulation, with a relationship which in winter has remained fairly steady through the 20th century. Notwithstanding this, there have been changes in the strength of the correlation between temperature and circulation. These fluctuations in climate-circulation relationships should be further investigated and addressed in studies of climate change, especially in the calibration of paleoclimatic time series and downscaling models.

A32A-0024 1330h POSTER

Diagnose of the Northern Hemisphere Annular Mode by Means of an Approach Based on the Relative Angular Momentum

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There is an interesting debate about the hemispheric character of the main mode of climate variability in the northern hemisphere extratropics (NAO-AO). Recent studies suggest that NAO is a regional expression of the Northern Hemisphere Annular Mode (AO). However the lack of an apparent link between Atlantic and Pacific sectors questions the annular paradigm. The main aim of this study is to diagnose the Northern Hemisphere annular mode using an annular magnitude, such as the relative angular momentum (RAM). Annual means of RAM are computed for the polar cap from 55 to 90°N using NCAR-NCEP and ECMWF reanalysis data. Values of RAM by 60° longitude sectors as well as the whole cap are then calculated and correlated to temperature and geopotential height at different levels. The results are similar in all cases: the Arctic region is dominated by very negative correlations whereas midlatitudes are dominated by positive correlations, although in this case there is not a regular annular pattern but intermittent significant regions. Since an annular pattern is characterized by its hemispheric symmetry, three symmetric latitude rings are expected in the correlation maps (negative-positive-negative), but only the polar one (negative) is detected, the rest being incomplete, so the results do not support the annular paradigm, understood as organized along latitude sectors, but an hemispheric dimension cannot be excluded.

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Anomalies in the Seasonal Cycle of Sea Level Pressure in Iceland and the North Atlantic Oscillation

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An analysis of a new homogenized time series of daily mean sea level pressure (SLP) from Reykjavik and Stykkishlukur Iceland from 1823-1999 is presented. Time series statistical techniques including harmonic analysis are used to identify the seasonal march of pressure and its variability through the record. The results are assessed regarding the North Atlantic Oscillation (NAO) index. In addition to the well-known annual cycle, the seasonal march of SLP has an appreciable semi-annual cycle, as well as anomalies (abrupt rises and