EAG Distinguished Lecture Tour 2013

Distinguished Lecturer:
Thomas Roeckmann

Lecture Abstracts

Lecture I: Oxygen isotope anomalies in the atmosphere

Isotope measurements belong to the most important tracers to study the Earth system. In atmospheric research, isotope effects are used to identify and quantify individual production and removal processes of atmospheric trace gases. For oxygen, with three stable isotopes, ^{16}O, ^{17}O and ^{18}O, measurements show that the variations between these three isotopes in the atmosphere are anomalous, also called "non-mass-dependent". The most important example for an anomalous oxygen isotope effect is the formation reaction of ozone (O_3).
At the same time, non-mass dependent oxygen isotope compositions have been detected in many other atmospheric molecules. In fact, there is almost no atmospheric species for which an anomaly has not been detected, or at least postulated. This is because O_3 is at the center of atmospheric oxidation reactions. Via chemical reactions the oxygen isotope anomaly is transferred from O_3 to other atmospheric compounds. Measuring the isotope anomaly therefore opens new opportunities to investigate chemical reaction mechanisms on the one hand, and mass fluxes between different species and reservoirs on the other hand. What started out as a peculiar isotope anomaly almost three decades ago has grown into a new research field that spans the full range from molecular physics studies to the reconstruction of climate parameters in the past.

Lecture II: Reconstructing changes in atmospheric trace gases in the industrial era from isotope measurements on air extracted from polar firn

In many cases, direct and comprehensive measurements on atmospheric composition are only available for the last 2-3 decades. To obtain information on changes before that time, air archives are needed, and polar ice cores are excellent archives of past atmospheric composition. Whereas available ice core records can go almost a million years back in time, air extracted from the firn, the top part of an ice sheet, can be used to cover the period of several decades to a century back in time. We have measured mixing ratio and isotopic composition of numerous trace gases on air samples extracted from firn at the NEEM ice core drilling site in Greenland. These data are combined with previously published measurements to reconstruct

consistent time series of isotope variations over the past 6 decades using a state of the art firn air model. It is important to take into account gravitational and diffusive fractionation in the firn air, which are in some cases of the same order as the actually isotope changes in the atmosphere. For example, for ^{13}C in methane (CH_4), reconstructions from individual sites are not always mutually consistent among the different sites, which emphasizes that the precise knowledge of the diffusive properties in the firn is very important. On the other hand, for nitrous oxide (N_2O) the isotope reconstructions between different sites agree well and allow reconstruction of a consistent isotope history that can be used to reconstruct the variations of the sources of N_2O in the past.

Lecture III: Isotopic variability of molecular hydrogen in the atmosphere and isotope signatures of its major sources

Molecular hydrogen (H_2) in the atmosphere has not been studied in great detail in the past because it does not directly affect the radiation balance. However, H_2 interacts with other atmospheric compounds via chemical reactions. The prospect of using H_2 as possible energy carrier in the future has led to renewed interest in understanding its atmospheric budget. Because of the large relative mass difference between hydrogen (H) and deuterium (D), isotope effects in molecular hydrogen (H_2) are particularly large and isotope measurements may provide additional information on the relative strength of the individual sources and sinks of H_2. Using an analytical system that enables isotope measurements on nanomolar quantities of H_2, we have investigated the isotopic composition of the different sources of H_2 in considerable detail. Hydrogen produced from photochemical processes is relatively enriched in deuterium, whereas hydrogen from combustion processes is depleted in D, and hydrogen formed in biological systems is extremely depleted in D.
In addition, in the framework of the European project EUROHYDROS, we have carried out regular isotope analyses on air samples collected at several sites worldwide in order to determine the spatial and temporal variability of the isotopic composition of H_2 in the atmosphere. In order to interpret these data, a full H_2 isotope scheme was implemented in the global model TM5. The results show that isotope measurements provide useful constraints on the global H_2 budget.

Lecture IV: The isotopic composition of long-lived trace gases in the stratosphere

Long-lived trace gases like nitrous oxide (N_2O) or methane (CH_4) reach the stratosphere where they are removed via photochemical reactions. These reactions are associated with isotope effects that usually enrich the trace gases in heavy isotopes. As part of the global atmospheric circulation, air that has been processed in the stratosphere returns to the troposphere and therefore the stratospheric isotope enrichments can considerably affect the tropospheric isotope budgets of

long-lived trace gases. We have investigated in detail the isotopic composition of several trace gases in the stratosphere, as well as the individual removal processes in laboratory experiments. Stratospheric samples were obtained during stratospheric balloon flights and on high altitude aircraft. The samples cover a large range in mixing ratios and show the largest heavy isotope enrichments observed in these compounds so far. In addition to the chemical removal, also dynamical processes affect the isotopic composition of trace gases in the stratosphere.