



Lagrangian Modeling of the Atmosphere



American Geophysical Union Chapman Conference

Grindelwald, Switzerland

9 -14 October 2011

AGU Chapman Conference on Lagrangian Modeling of the Atmosphere

Grindelwald, Switzerland

9 -14 October 2011

Conveners

John C. Lin, University of Waterloo, Canada

Dominik Brunner, Empa, Swiss Federal Laboratories for Materials Science and
Technology

Christoph Gerbig, Max Planck Institute for Biogeochemistry, Germany

Program Committee

Arlyn Andrews, NOAA Earth System Research Laboratory, USA

Massimo Cassiani, Norwegian Institute for Air Research

Roland Draxler, NOAA Air Resources Laboratory, USA

Kathy Law, LATMOS-CNRS, Université Pierre et Marie Curie, France

Paul Konopka, Forschungszentrum Jülich, Germany

Thomas Nehr Korn, Atmospheric and Environmental Research, USA

Viatcheslav Shershakov, State Institution “Research and Production Association
“Typhoon”, Russia

Petra Seibert, Institute of Meteorology, University of Natural Resources and Life
Sciences, Austria

Andreas Stohl, Norwegian Institute for Air Research

Barbara Stunder, NOAA Air Resources Laboratory, USA

David Thomson, United Kingdom Meteorological Office

Alex Vermeulen, Energy Research Centre of the Netherlands

Peter Webley, University of Alaska, Fairbanks, USA

Heini Wernli, ETH Zürich, Switzerland

John D. Wilson, University of Alberta, Canada

Gerhard Wotawa, Central Institute for Meteorology and Geodynamics, Vienna, Austria

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AGU Chapman Conference on Lagrangian Modeling of the Atmosphere

Meeting At A Glance

Sunday, 9 October 2011

1700h – 1900h Registration
1800h – 1900h Welcome Reception

Monday, 10 October 2011

0830h – 0850h Opening Remarks
0850h – 0910h History of Lagrangian Stochastic Model for Turbulent Dispersion
0910h – 1150h 1.1 Fundamental Theoretical Formulations
1010h – 1040h Coffee Break
0910h – 1200h 1.1 Fundamental Theoretical Formulations (cont.)
1210h – 1240h 2. Coupling Between Eulerian NWP/GCM and Lagrangian Models I
1240h – 1340h Lunch
1340h – 1500h 2. Coupling Between Eulerian NWP/GCM and Lagrangian Models II
1500h – 1530h Coffee Break
1530h – 1700h 1.3 Accounting for Uncertainties in Lagrangian Models
1700h – 1730h Discussion Topics 1 & 2
1730h – 1900h Dinner
1900h – 2100h Poster Session I

Tuesday, 11 October 2011

0820h – 1010h 8. Field Experiments and Observations to Test Lagrangian Models
1010h – 1030h Coffee Break and Preparation for Field Trip
1030h – 1730h Optional Excursion to Jungfrauoch (Tuesday)
1730h – 1900h Dinner
1900h – 2100h Poster Session II

Wednesday, 12 October 2011

0830h – 1020h 3. Application: Greenhouse Gases
1020h – 1050h Coffee Break
1050h – 1210h 3. Application: Greenhouse Gases (cont.)
1210h – 1240h 4. Application and Methods: Atmospheric Chemistry, Dispersion and
Mixing in the Troposphere and Planetary Boundary Layer I
1240h – 1340h Lunch
1340h – 1520h 4. Application and Methods: Atmospheric Chemistry, Dispersion and
Mixing in the Troposphere and Planetary Boundary Layer II
1520h – 1550h Coffee Break
1550h – 1720h 9. Application: Atmospheric Dynamics and Water Cycle
1720h – 1800h Discussion Topics 3, 4 & 9
1900h – 2100h Gala Dinner

Thursday, 13 October 2011

0830h – 1030h	5. Application: Processes in Upper Troposphere/Lower Stratosphere
1030h – 1050h	Coffee Break
1050h – 1150h	5. Application: Processes in Upper Troposphere/Lower Stratosphere (cont.)
1150h - 1240h	6. Application: Nuclear Releases I
1240h – 1340h	Lunch
1340h – 1440h	6. Application: Nuclear Releases II
1440h – 1530h	7. Application: Volcanic Ash, Dust, Aerosols
1530h – 1600h	Coffee Break
1600h – 1720h	7. Application: Volcanic Ash, Dust, Aerosols (cont.)
1720h – 1800h	Discussion Topics 5, 6 & 7
1800h – 1930h	Dinner
1930h – 2100h	Discussion and Closing Remarks

Friday, 14 October 2011

0800h – 1700h	Optional Excursion to Jungfraujoeh (Friday)
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SCIENTIFIC PROGRAM

SUNDAY, 9 OCTOBER

1700h – 1800h **Registration**

1800h – 1900h **Welcome Reception**

MONDAY, 10 OCTOBER

0830h – 0850h **Opening Remarks**

0850h – 0910h **History of the Lagrangian Stochastic Model for Turbulent Dispersion**

1.1 Fundamental Theoretical Formulations

Presiding: David Thomson, John D. Wilson

Eiger I & II

0910h – 0930h **James G. Esler** | A unified framework for global Eulerian and Lagrangian models of atmospheric transport

0930h – 0950h **Benjamin Devenish** | The geometry of turbulent dispersion

0950h – 1010h **Pasquale Franzese** | PDF Modeling of the Internal Fluctuations of a Tracer Release

1010h – 1040h Coffee Break

1040h – 1100h **Kenneth P. Bowman** | Lagrangian Diagnostics for Transport and Irreversible Stirring

1100h – 1120h **A. D. Kirwan** | The 3-D Lagrangian Description of Ocean Eddies

1120h – 1140h **Seoleun Shin** | Hamiltonian Particle-Mesh (HPM) methods for numerical modeling of atmospheric flows

1140h – 1200h **John D. Wilson** | Are “Rogue Velocities” in Lagrangian Stochastic Simulations Other Than a Manifestation of an Insufficiently Small Time Step?

2. Coupling Between Eulerian NWP/GCM and Lagrangian Models I

Presiding: Thomas Nehrkorn
Eiger I & II

1210h – 1240h **Sabine Brinkop** | Towards the development of a Lagrangian dynamical core (*INVITED*)

1240h – 1340h Lunch

2. Coupling Between Eulerian NWP/GCM and Lagrangian Models II

Presiding: Thomas Nehrkorn
Eiger I & II

1340h – 1400h **Thomas Nehrkorn** | Lagrangian Modeling of Greenhouse Gas Observations in Urban Environments

1400h – 1420h **Jennie L. Thomas** | Photochemical processing during long-range transport using Eulerian and Lagrangian approaches

1420h – 1440h **Glenn D. Carver** | Analysis of Aircraft Measurements of Atmospheric Composition by Eulerian and Lagrangian Models

1440h – 1510h Coffee Break

1.3 Accounting for Uncertainties in Lagrangian Models

Presiding: Christoph Gerbig, Petra Seibert
Eiger I & II

1530h – 1600h **Stefano Galmarini** | Multi-model model ensemble dispersion modelling: from empiricism to structure (*INVITED*)

1600h – 1620h **Andrew R. Jones** | Towards a probabilistic approach for hazard area assessment in dispersion modelling

1620h – 1640h **Heini Wernli** | The sensitivity of offline trajectory calculations on the temporal resolution of the meteorological input data

1640h – 1700h **Paul Konopka** | Diabatic versus kinematic trajectory modeling of stratospheric transport

1700h – 1730h Discussion Topics 1 & 2

1730h – 1900h **Dinner (Monday)**

1900h – 2100h **Poster Session I**
Eiger I & II

M-1 **John D. Wilson** | History of the Lagrangian Stochastic Model for Turbulent Dispersion, Part 1

- M-2 **Roland Draxler** | Data Archive of Tracer Experiments and Meteorology (DATEM)
- M-3 **Ayoe B. Hansen** | Test of Semi-Lagrangian Methods for Advection in the Danish Eulerian Hemispheric Model
- M-4 **Pirmin Kaufmann** | Sensitivity of wet deposition to the interpolation of meteorological fields in the offline coupled dispersion model Flexpart
- M-5 **Alexandre M. Ramos** | A new circulation type classification based upon Lagrangian air trajectories
- M-6 **Patrick Joeckel** | Lagrangian Transport in a Chemistry Climate Model: Methods and Applications
- M-7 **A. D. Kirwan** | A Lagrangian Perspective of the Deepwater Horizon Spill
- M-8 **José A. Orza** | The Influence of NAO on the Interannual Variability of Tropospheric Transport Pathways in Southern Europe
- M-9 **Dhanyalekshmi Pillai** | Towards regional carbon budgeting using WRF/STILT-VPRM: A comparison of Lagrangian and Eulerian models for atmospheric CO₂ transport
- M-10 **Roberto Kretschmer** | Uncertainty of simulated vertical mixing of CO₂ within the Lagrangian transport model framework WRF-STILT-VPRM
- M-11 **Michael Sprenger** | A Lagrangian climatology of orographic blocking
- M-12 **Michael Sprenger** | The Lagrangian Analysis Tool Lagranto
- M-13 **David Thomson** | History of the Lagrangian Stochastic model for turbulent dispersion, Part 2
- M-14 **Ana Maria Duran Quesada** | Role of the Caribbean Low Level Jet in the transport of moisture in the Intra Americas Sea region: A Lagrangian study based on backward trajectories
- M-15 **Paul Konopka** | Transport in the Chemical Lagrangian Model of the Stratosphere (CLaMS)
- M-16 **Peter Webley** | Improving the Accuracy of Eruption Source Parameters for Aviation Safety
- M-17 **Hanna Joos** | A Lagrangian climatology of warm conveyor belts over the past 20 years
- M-18 **Hanna Joos** | Microphysical processes and the associated latent heating along trajectories in an extra-tropical cyclone
- M-19 **Elena Novakovskaia** | Sensitivity of STILT footprints to assimilation of weather observations within the transport model
- M-20 **Michel S. Bourqui** | Validation of a new operational package for the Lagrangian diagnosis of stratosphere-troposphere exchange at Environment Canada

- M-21 **Kesavapillai Mohanakumar** | Exchange of Water Vapor from Troposphere to Stratosphere During Asian Summer Monsoon Season
- M-22 **Ingo Wohltmann** | The Lagrangian Chemistry and Transport Model ATLAS
- M-23 **Bojan Škerlak** | A global ERA-interim Climatology of Stratosphere-Troposphere Exchange and Identification of Key Physical Processes
- M-24 **Baerbel Vogel** | Using peroxyacetyl nitrate (PAN) - tracer correlations in the tropopause region to quantify stratosphere-troposphere exchange in Lagrangian model simulations
- M-25 **Brigitte Tschanz** | Analyzing Middle Atmospheric Water Vapor Variations during SSW 2010 using a Lagrangian Trajectory Model
- M-26 **Ignacio J. Pizzo** | Emission location dependent ozone depletion potentials for very short-lived halogenated species
- M-27 **Dominik Scheiben** | Lagrangian trajectory analysis of middle atmospheric ozone and water vapor during the sudden stratospheric warming of January 2010

TUESDAY, 11 OCTOBER

8. Field Experiments and Observations to Test Lagrangian Models

Presiding: Roland Draxler
Eiger I & II

- 0820h – 0850h **John D. Wilson** | Inverse Dispersion in Disturbed Surface Layer Flows Using a Three-Dimensional Backward Lagrangian Stochastic (‘3D-bLS) Model (*INVITED*)
- 0850h – 0910h **Arlyn E. Andrews** | Evaluation of Lagrangian Particle Dispersion Models using Radiocarbon and Carbon Dioxide Measurements
- 0910h – 0930h **Felix R. Vogel** | Potentials (and problems) of validating atmospheric transport and emission modeling of fossil fuel CO₂ and other tracers at an urban site by in-situ observations
- 0930h – 0950h **Michelle Cain** | A trajectory ensemble study of chemical and physical processes influencing ozone during transport over the North Atlantic
- 0950h – 1010h Coffee Break, Preparation for Field Trip
- 1030h – 1730h **Optional Excursion to Jungfrauoch - Tuesday**
1017h - Departure of train from railway station Grindelwald
1742h - Arrival back in Grindelwald

1730h – 1900h **Dinner (Tuesday)**

1900h – 2100h **Poster Session II**

Eiger I & II

- T-1 **Jerome Brioude** | A new inversion method to calculate emission inventories without a prior at mesoscale: Application to the anthropogenic CO₂ flux from Houston, Texas
- T-2 **John D. Wilson** | Application of the Backward Lagrangian Stochastic (bLS) Method to Quantify Methane Emission from a Lagoon
- T-3 **Zoë Fleming** | Untangling the seasonal and geographical influences at Cape Verde Atmospheric Observatory
- T-4 **Junming Wang** | PM₁₀ contribution from agricultural tilling operations
- T-5 **John C. Lin** | A backward-time Lagrangian air quality modelling system
- T-6 **Kae Tsunematsu** | Modeling transport of volcanic particles with a multiparticle cellular automata method
- T-7 **Chika Minejima** | Comparisons of observed and simulated CO₂, CO, CH₄, and O₂ at Hateruma Island, JAPAN, by using FLEXPART and global coupled Eulerian-Lagrangian transport model
- T-8 **Peter Bedwell** | Cloud gamma modelling in the UK Meteorological Office's NAME III Model
- T-9 **Mikhail A. Novitsky** | The Lagrange Model of an Accidental Release Transport in an Atmosphere with Limitation of Cross-section Component of a Wind Speed Fluctuation
- T-10 **Peter W. Webley** | Pre-eruption warnings of the potential hazards from volcanic ash clouds
- T-11 **Tamir G. Reisin** | Numerical Simulations of Cloud Rise and Cloud Top Determination Following an Explosion Using a Lagrangian Model
- T-12 **Helen N. Webster** | Forecasting Peak Ash Concentrations within the Volcanic Cloud from the 2010 Eruption of Eyjafjallajökull, Iceland
- T-13 **Sabine Eckhardt** | The impact of North American and Asian emissions on carbon monoxide and ozone concentrations over Europe
- T-14 **Réal D'Amours** | Application of Lagrangian modelling to radioactive releases following the Fukushima nuclear accident
- T-15 **Jooil Kim** | Measurements of Greenhouse Gases at Gosan (Jeju Island, Korea) for Regional Analysis of Emissions in East Asia
- T-16 **Christoph Keller** | Inverse Modeling of European Halogenated Greenhouse Gas Emissions Based on Atmospheric Observations and the Lagrangian Particle Dispersion Model FLEXPART

- T-17 **Christina Schnadt Poberaj** | Understanding Recent Methane Growth Rate Variability Using a Global Lagrangian Transport Model – First Results
- T-18 **Maria Cabello** | Influence of interannual variability of transport on surface based measurements of greenhouse gases
- T-19 **Joshua S. Benmergui** | Parameterizing atmospheric chemistry dependent on contact with the Earth’s surface using a time-reversed Lagrangian particle model (STILT)
- T-20 **Nitin K. Jaiswal** | Particulate Source Apportionment in Central India using Hybrid Single-Particle Lagrangian Integrated Trajectory Model
- T-21 **Franziska Aemisegger** | Lagrangian investigation of high-frequency variations in stable isotope composition of atmospheric boundary layer water vapour
- T-22 **Heini Wernli** | A Lagrangian method to identify regions potentially affected by wet deposition of radioactive emissions from the Fukushima power plant - a climatological study
- T-23 **Alexander T. Vermeulen** | Spatial Estimates for Methane and Nitrous Oxide Emissions at the Continental Scale Using a Direct Inversion Technique With Recursive Source Area Aggregation
- T-24 **Kuoying Wang** | Assessment of industrial air pollution episodes with high resolution Lagrangian model
- T-25 **Oscar J. Guerrero** | Assessing wetland and anthropogenic methane fluxes in Colombia and Panama from Lagrangian simulation analysis of aircraft-borne measurements during TC4
- T-26 **Alex White** | Source sensitivity in surface-to-stratosphere transport addressed using an Eulerian and a Lagrangian adjoint model

WEDNESDAY, 12 OCTOBER

3. Application: Greenhouse Gases

Presiding: Arlyn E. Andrews, Alexander T. Vermeulen
Eiger I & II

- 0830h – 0900h **Andreas Stohl** | Source identification and quantification with Lagrangian model output: strategies with different complexity (*INVITED*)
- 0900h – 0920h **Christoph Gerbig** | Assessment of Network design for greenhouse gas observing systems using Lagrangian footprint simulations
- 0920h – 0940h **Ute Karstens** | Regional-scale atmospheric inversions of greenhouse gas fluxes in Europe

- 0940h – 1000h **Scot M. Miller** | Constraints on nitrous oxide and methane sources over North America and their underlying drivers
- 1000h – 1020h **Ignacio J. Pizzo** | Constraints on CO₂ flux emissions based on reconstructions of in-situ measurements from Lagrangian stochastic inversion
- 1020h – 1050h Coffee Break
- 1050h – 1110h **John B. Miller** | Forward and Inverse Modeling of CO₂ and δ¹³CO₂ at North American Tall Towers using the FLEXPART Lagrangian Model
- 1110h – 1130h **Huilin Chen** | Simulations of carbonyl sulfide (COS) and CO₂ for North America using STILT
- 1130h – 1150h **Dominik Brunner** | Regional methane sources observed from a motor glider and simulated with a Lagrangian particle dispersion model at kilometre scale resolution
- 1150h – 1210h **Janusz Eluszkiewicz** | Lagrangian Modeling of Satellite Greenhouse Gas Observations

4. Application and Methods: Atmospheric Chemistry, Dispersion and Mixing in the Troposphere and Planetary Boundary Layer I

Eiger I & II

- 1210h – 1240h **Alexander T. Archibald** | Simulating chemistry using a hybrid approach in a Lagrangian particle dispersion model: Impacts of biogenic emissions and gas phase chemical mechanism (*INVITED*)
- 1240h – 1340h Lunch

4. Application and Methods: Atmospheric Chemistry, Dispersion and Mixing in the Troposphere and Planetary Boundary Layer II

Presiding: Massimo Cassiani, Kathy Law

Eiger I & II

- 1340h – 1400h **Massimo Cassiani** | A novel approach to model concentration fluctuations and chemical reactions in a dispersing plume based on Lagrangian particle and particle-grid methods
- 1400h – 1420h **Raymond W. Arritt** | Biology Meets Turbulence: Lagrangian Modeling of Pollen Dispersion and Viability
- 1420h – 1440h **Enrico Ferrero** | Beyond the Limits of the Lagrangian Particle Models, From the Chemistry to the Entrainment
- 1440h – 1500h **Stephan Henne** | Deposition of trifluoroacetic acid (TFA) resulting from future emissions of HFO-1234yf from mobile air conditioners in Europe

1500h – 1520h **Sandy Ubl** | Exploring Atmospheric Transport of PCBs Measured in the Arctic

1520h – 1550h Coffee Break

9. Application: Atmospheric Dynamics and Water Cycle

Presiding: Heini Wernli

Eiger I & II

1550h – 1620h **Harald Sodemann** | An intercomparison of Lagrangian methods to diagnose evaporation regions of water vapour and precipitation (*INVITED*)

1620h – 1640h **Ana Maria Duran Quesada** | Variability of sources of moisture in the Intra Americas Sea region based on a Lagrangian approach

1640h – 1700h **Margarida L. Liberato** | The Role of Extratropical Cyclones on the Transport of Atmospheric Water Vapour

1700h – 1720h **Annette K. Miltenberger** | Lagrangian Perspective on Warm-Rain Orographic Precipitation in Idealized Three-Dimensional Simulations

1720h – 1800h Discussion Topics 3, 4 & 9

1900h – 2100h **Gala Banquet**

THURSDAY, 13 OCTOBER

5. Application: Processes in Upper Troposphere/Lower Stratosphere

Presiding: Paul Konopka

Eiger I & II

0830h – 0900h **Laura Pan** | Chemical Transport and Mixing near the Tropopause: Integrated Studies using Chemical Tracer Measurements and Lagrangian Models (*INVITED*)

0900h – 0925h **Bernard Legras** | Lagrangian Transport in the Tropical Tropopause Layer and Convective Sources (*INVITED*)

0925h – 0950h **Michael Sprenger** | Lagrangian perspective of stratosphere-troposphere exchange - physical processes associated with the crossing of the extra-tropical tropopause (*INVITED*)

0950h – 1010h **Michel S. Bourqui** | A one-year global high-resolution Lagrangian climatology of stratosphere-troposphere exchange

1010h – 1030h **Anne Kunz** | Atmospheric transport pathways near the jet streams and their impact on the UT/LS trace gas distribution

- 1030h – 1050h Coffee Break
- 1050h – 1110h **Peter Haynes** | The advection-condensation paradigm for stratospheric water vapour
- 1110h – 1130h **Peter M. Hoor** | Transport Times in the ExUTLS: Does Static Stability Matter?
- 1130h – 1150h **John W. Bergman** | Seasonal contrasts of transport of convective outflow upward through the Tropical Tropopause Layer

6. Application: Nuclear Releases I

Presiding: Viatcheslav Shershakov, Gerhard Wotawa
Eiger I & II

- 1150h – 1220h **Dèlia Arnold** | flexRISK: Lagrangian particle dispersion modelling for the assessment of nuclear risks in Europe (*INVITED*)
- 1220h – 1240h **Jiye Zeng** | Introducing an Implementation of Parallel Computing for Lagrangian Modelling of Particle Dispersion in the Atmosphere
- 1240h – 1340h Lunch

6. Application: Nuclear Releases II

Presiding: Viatcheslav Shershakov, Gerhard Wotawa
Eiger I & II

- 1340h – 1400h **Mykola Talerko** | Evaluation of Potential and Limitations of the Lagrangian-Eulerian Mesoscale Model for the Reconstruction of Space-Time Features of Contamination Fields in Belarus and Ukraine Caused by the Chernobyl Accident
- 1400h – 1420h **Viatcheslav Shershakov** | The model STADIUM as an instrument for estimating radioactivity dispersion in case of a nuclear accident
- 1420h – 1440h **Gerhard Wotawa** | Simulation of the transport of radioactivity from the Fukushima nuclear accident

7. Application: Volcanic Ash, Dust, Aerosols

Presiding: Peter W. Webley
Eiger I & II

- 1440h – 1510h **Helen Dacre** | Evaluating the structure and magnitude of the ash plume during the initial phase of the 2010 Eyjafjallajökull eruption using lidar observations and NAME simulations (*INVITED*)
- 1510h – 1530h **Roger P. Denlinger** | Objective and quantitative comparison of volcanic ash dispersal from different transport models
- 1530h – 1600h Coffee Break

- 1600h – 1620h **Imogen P. Heard** | A Comparison of Atmospheric Dispersion Model Predictions with Observations of SO₂ and Sulphate Aerosol from Volcanic Eruptions
- 1620h – 1640h **Alain Malo** | Application of the Canadian Atmospheric Lagrangian Particle Transport and Dispersion Model MLDP0 to the May-June 2010 Forest Fires Episode in Central Québec: A Case Study for Quantitative Validations
- 1640h – 1700h **Luca Mortarini** | Lagrangian Particle Model Approach to Two-Phase Releases. The MicroSpray New Module
- 1700h – 1720h **Jesús A. Hernández** | EDA s Classifier Systems for Identifying Patterns of Particulate Matter (PM_x) Dispersion in a Study Area
- 1720h – 1800h Discussion Topics 5, 6 & 7
- 1800h – 1930h **Dinner (Thursday)**
- 1930h – 2100h **Discussion and Closing Remarks**

FRIDAY, 14 OCTOBER

- 0800h – 1700h **Optional Excursion to Jungfraujoch - Friday**
0847h - Departuren of train from railway station Grindelwald
1612h - Arrival back in Grindelwald

ABSTRACTS

listed by name of presenter

Aemisegger, Franziska

Lagrangian investigation of high-frequency variations in stable isotope composition of atmospheric boundary layer water vapour

Aemisegger, Franziska¹; Pfahl, Stephan¹; Sodemann, Harald¹; Wernli, Heini¹

1. Institute of Atmospheric and Climate Sciences, ETHZ, Zürich, Switzerland

Stable water isotopes can be regarded as naturally available tracers of phase changes in the atmosphere. The differences in volatility and diffusivity of the isotopologues lead to isotopic fractionation during phase transitions, which depends on environmental conditions, like temperature, relative humidity and wind velocity. Stable water isotopes can thus give us some hints on important moist atmospheric mechanisms like cloud formation, evaporation and transpiration at the land surface. Until recently measurements of stable water isotopes were possible only at relatively low temporal resolution using expensive mass spectroscopic techniques. At these temporal scales, variability in isotopic composition of meteoric waters reflects the behaviour of the climatic controls on evaporation and transport patterns. In this work, we used a novel laser spectroscopic instrument that can provide precise stable water isotopic measurements at high temporal resolution (<1 hour). This allows us to perform process-based investigations of the atmospheric water cycle at the time scales of significant weather events. Since the water isotopic composition of an air parcel is determined by the integrated history of phase changes and mixing processes from evaporation to the point of measurement, we adopted a Lagrangian perspective to analyse the data. We applied a moisture source and source condition identification algorithm from 3D kinematic backward trajectories based on atmospheric analysis data. Here, we present the results from a summer measurement campaign in a prealpine region in Switzerland. The diagnosed sources in this case are local, with an average distance to the moisture source of 150 km. Strong regional moisture recycling is found to dominate the isotope signal, which predominantly reflects boundary layer dynamics. The correlation between temperature at the source and the measured isotopic composition is weaker than in previous studies, which are based on daily data. Different hypotheses are formulated to explain this finding.

Andrews, Arlyn E.

Evaluation of Lagrangian Particle Dispersion Models using Radiocarbon and Carbon Dioxide Measurements

Andrews, Arlyn E.¹; Petron, Gabrielle^{2, 1}; Trudeau, Michael^{3, 1}; Chen, Huilin¹; Eluszkiewicz, Janusz⁴; Nehkorn, Thomas⁴; Henderson, John⁴; Draxler, Roland⁵; Stein, Ariel^{11, 5}; Lin, John⁷; Wen, Deyong⁷; Gerbig, Christoph⁸; Uliasz, Marek¹³; Schuh, Andrew¹³; Miller, John^{2, 1}; Lehman, Scott⁶; Guilderson, Tom⁹; LaFranchi, Brian⁹; Sweeney, Colm^{2, 1}; Karion, Anna^{2, 1}; Kofler, Jonathan^{2, 1}; Fischer, Marc¹²; Gurney, Kevin¹⁰

1. NOAA Earth System Research Laboratory, Boulder, CO, USA
2. Cooperative Institute for Research in Environmental Sciences, University of Colorado, Boulder, CO, USA
3. Cooperative Institute for Research in the Atmosphere, Colorado State University, Fort Collins, CO, USA
4. Atmospheric and Environmental Research, Inc., Lexington, MA, USA
5. NOAA Air Resources Laboratory, Silver Spring, MD, USA
6. Institute of Arctic and Alpine Research, University of Colorado, Boulder, CO, USA
7. University of Waterloo, Waterloo, ON, Canada
8. Max-Planck-Institut für Biogeochemie, Jena, Germany
9. Lawrence Livermore National Laboratory, Livermore, CA, USA
10. Arizona State University, Tempe, AZ, USA
11. Resources Technology, Inc., Laurel, MD, USA
12. Lawrence Berkeley National Laboratory, Berkeley, CA, USA
13. Colorado State University, Fort Collins, CO, USA

Lagrangian Particle Dispersion Models (LPDMs) are useful tools for diagnosing upwind sources and sinks that affect greenhouse gas and air pollutant distributions, and model-data and model-model comparisons can provide information about uncertainty in transport formulations. Different LPDMs have different parameterizations and can be driven by a variety of meteorological datasets, including operationally available products like NARR, ECMWF, and NAM, or customized simulations using models like WRF, RAMS or BRAMS. Customized simulations can vary in resolution, physics, assimilation schemes and available output. We have selected two benchmark cases that are relevant for greenhouse gas monitoring and air pollution studies. (1) We convolve the Vulcan inventory of CO₂ emissions with LPDM influence functions and compare the resulting modeled fossil fuel CO₂ signal with radiocarbon-based estimates. Radiocarbon data is now available from several tall tower, mountaintop and aircraft sites in North America. The atmospheric distribution of radiocarbon is affected primarily by fossil fuel combustion, and inventory-based estimates of fossil fuel CO₂ emissions are thought to

be reasonably accurate in the aggregate (of order 10% uncertainty), although the temporal and spatial patterns are less well known. We plan to compare modeled and observed radiocarbon for different seasons and different heights in the atmosphere. (2) We will also evaluate daytime modeled and observed CO₂ concentrations and vertical gradients, where biological flux estimates are taken from the CarbonTracker data assimilation system. Biological carbon dioxide flux estimates have large uncertainty and exhibit complex spatial and temporal patterns. Although “truth” is unknown, model comparisons using the same flux fields can provide insight into how differences in model transport may affect flux estimates. We hope these model comparisons and model-data comparisons will contribute to a more comprehensive and community-wide evaluation of LPDMs used to estimate greenhouse gas and pollutant emissions. An important aspect of this work is identifying useful metrics to quantitatively describe model differences.

Archibald, Alexander T.

Simulating chemistry using a hybrid approach in a Lagrangian particle dispersion model: Impacts of biogenic emissions and gas phase chemical mechanism (*INVITED*)

Archibald, Alexander T.^{1,2}; Redington, Alison³; Witham, Claire³; Shallcross, Dudley²; Pyle, John¹; Manning, Alistair³

1. National Centre for Atmospheric Science, University of Cambridge, Cambridge, United Kingdom
2. School of Chemistry, University of Bristol, Bristol, United Kingdom
3. Atmospheric Dispersion Group, Met Office, Exeter, United Kingdom

Air quality is of primary importance in both rural and urban areas. However, our ability to forecast air pollution and understand pollutant control strategies relies heavily on numerical models. Essentially two frameworks exist for modelling air quality - the Lagrangian framework and the Eulerian framework. Over the past decade or so the Eulerian has been adopted more frequently for forecasting and analysing air pollution episodes than the Lagrangian model. However, with the advent of modern computing it is possible to combine both methods to create hybrid air quality models that take into account the benefits of both modelling approaches. In this work we describe how chemistry is included in the UK Met Office Numerical Atmospheric-dispersion Modelling Environment (NAME), a Lagrangian particle dispersion model, and show results comparing the model to observations of air pollutants from intensive field campaigns focusing on urban (PUMA) and rural (TORCH) air quality in the United Kingdom. The importance of biogenic emissions is tested as is the choice of chemical mechanism. By performing simulations with a chemical mechanism of intermediate complexity (200 species and 600 reactions) it is found that uncertainties in emissions remain a key area in correctly simulating ground level air quality but gas phase mechanism choice is a key component for air quality modelling.

Arnold, Dèlia

flexRISK: Lagrangian particle dispersion modelling for the assessment of nuclear risks in Europe (*INVITED*)

Arnold, Dèlia¹; Seibert, Petra¹; Kromp-Kolb, Helga¹; Gufler, Klaus²; Arnold, Nikolaus²; Kromp, Wolfgang²; Wensch, Antonia³; Mraz, Gabriele³; Sutter, Philipp³

1. Institute of Meteorology, University of Natural Resources and Life Sciences, Vienna, Vienna, Austria
2. Institute of Safety and Risk Sciences, University of Natural Resources and Life Sciences, Vienna, Austria
3. Austrian Institute of Ecology, Vienna, Austria

The flexRISK project studies the geographical distribution of the consequences of severe accidents in nuclear facilities in Europe with a flexible set of tools. The dispersion of radioactivity released in hypothetical severe accidents at 90 nuclear sites has been simulated for a large number of meteorological situations. The assumed releases are evenly distributed over seasons and time of day during the period 2000-2009, plus 90 cases in 1995, considered as a climatologically representative year. This results in a set of 2720+90 meteorological situations. For each of them, dispersion calculations have been carried out for all the considered nuclear sites and with several release shapes (one or more release phases and release heights, to cover selected accidents specific to each reactor on a site). The total number of runs is about 500,000. The simulations were carried out in forward mode with a slightly modified version of the Lagrangian particle dispersion model FLEXPART 8.2 and meteorological input from the ERA-Interim data set. Modifications were implemented to reduce the mass storage and CPU time needs. Dry and wet deposition fields were added into one single field. Deposition fields are output as incremental instead of being accumulated. Runs are stopped when the mass remaining airborne within the domain goes below a threshold. The set-up of the runs (number of particles etc.) was tuned to fit into the total CPU time allotted on the Vienna Scientific Cluster (VSC) and for the output to fit into the RAID disk space reserved. This finally resulted in a large 1x1 deg output domain stretching from the North Cape to Northern Africa and the Persian Gulf, and a smaller output domain with much higher resolution (ca. 10x10 km²), covering Europe without Russia and northern Scandinavia. As a result of the required trade-off, the number of computational species had to be limited to two: noble gas and aerosol. For each of these species, the air concentration and the ground deposition was obtained as a function of time, yielding about 2.5 TB of compressed data. A scripting environment in bash and python has been set up to handle the vast number of calculations with associated input and output files. For a selected set of stations and dates, a first visualisation of results has been published online for I-131 and Cs-137. One of the findings relevant for future work is that the 1 deg output is clearly too coarse, even after days of transport, whereas the 10 km output suffers from insufficient particle numbers after a certain time. The fact that particle number is determining the

effective resolution and not a fixed grid is the big advantage of Lagrangian models. However, the issues related to conversion to gridded output deserve more attention for future improvements. Acknowledgment: This work is funded by the Austrian Climate and Energy Fund (KLI.EN), programme Neue Energien 2020. FLEXPART calculations were performed in the VSC with kind support of its staff.

<http://flexrisk.boku.ac.at/>

Arritt, Raymond W.

Biology Meets Turbulence: Lagrangian Modeling of Pollen Dispersion and Viability

Arritt, Raymond W.¹; Viner, Brian J.¹; Westgate, Mark E.¹

1. Department of Agronomy, Iowa State University, Ames, IA, USA

Adoption of genetically modified (GM) crops has raised concerns that GM traits can accidentally cross into conventional crops or wild relatives through the transport of wind-borne pollen. In some cases, such as when plant-made pharmaceuticals (PMPs) are being produced, there is concern over even small probabilities of outcrossing. In assessing this risk it is necessary not only to compute the transport and dispersion of pollen but also to account for the fact that pollen is a living organism (gametophyte) whose viability is closely linked to its moisture content; i.e., pollen grains become non-viable as they lose moisture. The Lagrangian approach is particularly well suited to this challenge because it permits diagnosis of environmental temperature and moisture that pollen grains experience as they travel. We taken advantage of the Lagrangian method by combining a high-resolution version of the WRF meteorological model with a Lagrangian particle dispersion model to predict maize pollen dispersion and viability. WRF is used to obtain fields of wind, turbulence kinetic energy, temperature, and humidity archived at one-minute intervals which are then used as input to the Lagrangian dispersion model. The dispersion model in turn predicts transport of a statistical sample of a pollen cloud from source plants to receptors. We use the three-dimensional temperature and moisture fields from WRF to diagnose vapor pressure deficit at each point along the path of each tracer particle (virtual pollen grain), from which changes in moisture content of the pollen grains and consequent loss of viability are calculated. We tracked the movement and viability of approximately 100×10^6 particles released near the surface. Small amounts of pollen were transported 5 km or more from the source. Previous estimates of maize pollen transport using only surface conditions have predicted transport only to a few tens of meters, owing to the large terminal velocity of maize pollen grains (about 20-30 cm s⁻¹). Conversely, we found that turbulent motions in the convective boundary layer counteract the large terminal velocity of maize pollen grains and lift them to heights of several hundred meters, so that they can be transported long distances before settling to the ground. We also found that pollen lifted into the upper part of the boundary layer remains more viable than has been inferred from surface

observations of temperature and humidity. This is attributed to the thermal and moisture structure that typifies the daytime atmospheric boundary layer. Temperature decreases with height approximately as the dry adiabatic lapse rate while absolute humidity remains approximately constant, producing an environment of low vapor pressure deficit in the upper boundary layer which helps preserve the viability of pollen. Our results illustrate the complex interplay between transport and biology, and are consistent with recent studies using both piloted and remote-controlled aircraft that have shown the presence of viable maize pollen through the entire depth of the convective boundary layer. We recommend that field experiments be carried to assess the transport of fugitive pollen to long distances.

Bedwell, Peter

Cloud gamma modelling in the UK Meteorological Office's NAME III Model

Bedwell, Peter¹; Wellings, Joseph¹

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The Health Protection Agency performs a broad spread of radiological exposure assessments to develop and support the provision of health protection advice. Such assessments include investigating the impact of discharges to the environment from controlled releases to support decisions on authorisation and regulatory control, and investigating the consequences of accidental releases for both emergency planning and response. Atmospheric releases are commonly considered in radiological exposure assessments and result in a wide range of potential routes of exposure of an individual or a population. One such route, external exposure to gamma rays irradiating directly from a dispersing plume, may contribute significantly to dose if the radionuclides discharged are strong gamma emitters or are noble gases (which do not deposit, thus limiting the number of potential exposure pathways). The nature of cloud gamma modelling necessitates full integration within a dispersion model, unlike other exposure pathways which may be considered subsequent to the dispersion modelling. The inclusion of the ability to estimate cloud gamma dose in the Met Office's Lagrangian Particle Dispersion model, NAME III, is necessary for use in comprehensive assessments of the consequences and risks of a radiological release to atmosphere. This paper describes the cloud gamma modelling approaches implemented within NAME III and a method for integrating the approaches. Recent work performed to validate the model against measurement data are described. The paper concludes by considering the potential applications and future work.

Benmergui, Joshua S.

Parameterizing atmospheric chemistry dependent on contact with the Earth's surface using a time-reversed Lagrangian particle model (STILT)

Benmergui, Joshua S.¹; Lin, John C.¹; Sangeeta, Sharma²

1. Earth and Environmental Sciences, University of Waterloo, Waterloo, ON, Canada
2. Climate chemistry Measurements and Research, Environment Canada, Toronto, ON, Canada

Much atmospheric chemistry that occurs in the Arctic areas around the ice edge and refreezing leads depend on the air-mass contact with sea ice. The processes involved can be direct fluxes of the constituent in question for compounds such as natural sulfur aerosols, or the flux of a reagent that affects the concentration of another constituent as is the case for the depletion of gaseous elemental mercury. The Footprint function in the Stochastic Time Inverted Lagrangian Transport Model provides a measure of air-mass contact with the Earth's surface. In this work we create parameterizations of surface fluxes of gaseous elemental mercury and methanesulfonic acid measured at the N. B. A. Trivett Global Atmospheric Watch Lab in Alert, Canada which include projections of the footprint onto a map of the Earth's surface, categorized into areas important to the flux. To create these categories we use sea ice concentrations data provided by the National Sea Ice Data Center and Ice age data provided by the University of Colorado. Other parameters such as temperature and solar radiation measured at Alert which are important to the chemical processes are also included. The parameters include tunable coefficients that are adjusted to create the best fit with historical data. After finding the best parameters which serve as input for the parameterization, we attempt to use it to determine the flux of the constituent along the pathways of air-mass trajectories.

Bergman, John W.

Seasonal contrasts of transport of convective outflow upward through the Tropical Tropopause Layer

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Winter-summer contrasts of the transport of air from the boundary layer to the lower stratosphere at low latitudes are investigated with ensembles of backward-trajectory calculations that track parcels from the 380 K isentropic surface to their convective detrainment into the tropical tropopause layer. Results are consistent with known properties of the TTL, convective activity, and previous trajectory studies. In particular, weak upwelling in the TTL during boreal summer compared to winter both slows the

ascent of parcels through the TTL and increases the convective threshold for that ascent, which severely restricts the abundance of young air in the stratosphere during summer. This, along with the Asian anticyclone, also makes monsoon-related convection over the Indian subcontinent the dominant source of young air during summer. In contrast, winter sources are spread over the southern continents and the W. Pacific. This contrast in the location of convective sources has strong implications for the stratospheric abundance of species that enter the atmosphere at the surface in concentrations that are geographically dependent. Sensitivity tests are used to expose weaknesses of the calculation and reveal important dynamical influences on the efficiency of surface-to-stratospheric transport. The most interesting of these results indicate an important role of anti-cyclonic circulations during winter as well as summer and that our results are not sensitive to small-scale perturbations. The latter has the encouraging implication that these calculations depend most on features of the circulation that are the most robust. However, uncertainties in vertical velocity and convective detrainment efficiency still place large constraints on our ability to draw definitive conclusions.

Bourqui, Michel S.

Validation of a new operational package for the Lagrangian diagnosis of stratosphere-troposphere exchange at Environment Canada

Bourqui, Michel S.¹; Yamamoto, Ayako¹; Tarasick, David²; Moran, Michael²; Argall, Stephen³; Beaudoin, Louis-Philippe⁴; Beres, Ildiko²; Davies, Jonathan²; Elford, Andrew²; Guertin, Chad²; Hocking, Wayne³; Osman, Mohammed³; Wilkinson, Ronald⁴

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2. Environment Canada, Downsview, ON, Canada
3. University of Western Ontario, London, ON, Canada
4. Canadian Space Agency, St-Hubert, QC, Canada

A new operational diagnostic package for stratosphere-troposphere exchange (STE) has been developed for Environment Canada (EC). The STE calculations are made daily on the basis of EC global forecasts (0.3x0.3 degrees x 80 levels up to 0.1hPa x 1hour). Following a Lagrangian approach, 18 million trajectories are calculated for 48h from five different times (0, 24, 48, 72, 96 hours) along each global forecast, starting from a global grid spanning the atmosphere from 600hPa to 10hPa. The trajectories crossing the 2PVU dynamical tropopause or the 380K isentrope are then selected and extended for four days forward or backward. A number of diagnostics are calculated, including global maps of the mass flux across the 2PVU tropopause and the 380K isentrope, and clusters of cross-tropopause transport events reaching the lower troposphere (700hPa). This data set is global and will shed new insights into STE processes, their frequency of occurrence in different regions of the world, and their possible impacts on the chemical composition of the troposphere and the lower stratosphere, and on surface air quality. It will also be useful as a new

reference for validating cross-tropopause fluxes in general circulation models or in chemistry transport models. This real-time forecasting of STE was used extensively in a recent observational campaign focussing on STE with balloon sondes launched daily from Montreal (QC, CA), Egbert and Walsingham (ON, CA) through the month of July 2010. This talk will present a thorough validation of this new high resolution Lagrangian STE forecast data against these balloon sonde measurements. The results are very encouraging, with an overall good match between observations and STE forecasts, but with a tendency for the STE forecasts to underestimate the frequency of deep stratospheric intrusions.

<http://www.meteo.mcgill.ca/bourqui/>

Bourqui, Michel S.

A one-year global high-resolution Lagrangian climatology of stratosphere-troposphere exchange

Bourqui, Michel S.¹; Paull, Georgina¹; Yamamoto, Ayako¹; Son, Seok-Woo¹; Straub, David¹; Tarasick, David²; Moran, Michael²

1. McGill University, Montreal, QC, Canada
2. Environment Canada, Downsview, ON, Canada

A new Lagrangian diagnostic package for stratosphere-troposphere exchange (STE) has been operational at Environment Canada (EC) since July 2010. The STE calculations are made daily on the basis of EC global forecasts (0.3x0.3 degrees x 80 levels up to 0.1hPa x 1hour). Following a Lagrangian approach, 18 million trajectories are calculated for 48h from five different times (0, 24, 48, 72, 96 hours) along each global forecast, starting from a global grid spanning the atmosphere from 600hPa to 10hPa. The trajectories crossing the 2PVU dynamical tropopause or the 380K isentrope are then selected and extended for four days forward or backward. A number of diagnostics are calculated, including global maps of the mass flux across the 2PVU tropopause and the 380K isentrope, and clusters of cross-tropopause transport events reaching the lower troposphere (700hPa). In this talk, we will discuss the results of the first year of operation of these calculations. Detailed geographical maps and seasonal cycles of the mass flux across the dynamical tropopause will be presented. An analysis of deep stratospheric intrusions (defined as stratospheric air crossing 700hPa) will also be presented. These results will be discussed in comparison with past published studies.

<http://www.meteo.mcgill.ca/bourqui/>

Bowman, Kenneth P.

Lagrangian Diagnostics for Transport and Irreversible Stirring

Bowman, Kenneth P.¹

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Lagrangian methods provide powerful tools for qualitative and quantitative understanding of transport and

irreversible stirring in a wide range of fluid flows. This talk will review some methods from dynamical systems theory that can be used to map the geometric structure of transport, describe numerical tools to implement these methods for observed fluid flows, and demonstrate their application to large-scale atmospheric flows. Lagrangian calculations using assimilated winds are compared with high-resolution in situ measurements of trace constituents. The methods discussed can be used both with retrospective analyses for diagnostic purposes and with forecast winds for purposes such as planning for research aircraft flights.

<http://atmo.tamu.edu/profile/KBowman>

Brinkop, Sabine

Towards the development of a Lagrangian dynamical core (*INVITED*)

Brinkop, Sabine¹; Grewe, Volker¹; Joeckel, Patrick¹; Sausen, Robert¹; Yserentant, Harry²; Shin, Seoleun³; Reich, Sebastian³

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3. Institut fuer Mathematik, Universitaet Potsdam, Potsdam, Germany

Most global atmosphere models with Lagrangian transport schemes are up to now hybrid models, for instance, the chemistry climate model EMAC with the Lagrangian tracer transport scheme ATTILA. EMAC optionally uses the Lagrangian model ATTILA for the transport of trace species, but is based on a spectral dynamical core for the calculation of the wind field. This inconsistency poses a major drawback since the advantages of the Lagrangian approach cannot be fully exploited. Within the DFG-project METSTRÖM a Lagrangian dynamical core is under development. This will lead to a consistent description of the Lagrangian mass field and the corresponding wind field, which forces the movement of the parcels. Two methods are tested: 1. The Finite-Mass Method (FMM). 2. The Hamiltonian Particle Mesh (HPM) method. The Lagrangian parcels within EMAC/ATTILA have only a mass but no volume and are thus regarded as centroids. Using the Finite-Mass Method we add a volume to each parcel. The Finite-Mass method divides the atmosphere into small mass packets, each having certain degrees of freedom. The parcels move under the influence of inner and outer forces according to the laws of thermodynamics and can intersect each other. These parcels dynamically adapt in form, size and orientation to the local flow field. For a climate model we have to implement the Finite-Mass method on a sphere, which is a new development. Therefore the Finite-Mass method is at first implemented into the EMAC/ATTILA model without any feedback, such that the Lagrangian parcels move only under the influence of an external velocity field. Several sensitivity simulations with respect to parcel behaviour, development of parcel volume, shape and associated statistics have been performed. Results

of these tests will be presented. In the second approach, the HPM method, the atmosphere mass is represented by centroids (like in ATTILA) without volume. The Lagrangian mass distribution is mapped to the model grid using a tensor product cubic b-spline function. The resultant mass distribution is smoothed and then used to calculate the pressure field, from which the spectral core of EMAC calculates the wind. First results will be shown.

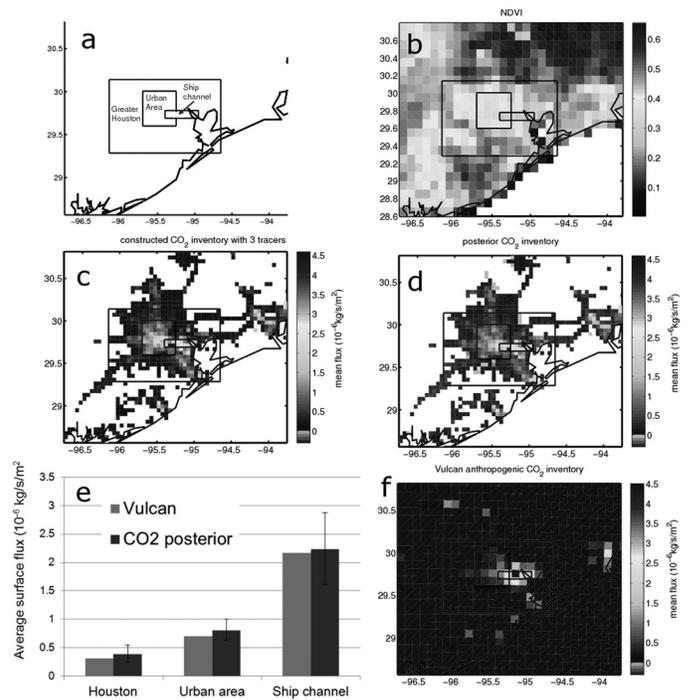
Brioude, Jerome

A new inversion method to calculate emission inventories without a prior at mesoscale: Application to the anthropogenic CO₂ flux from Houston, Texas

Brioude, Jerome^{1,2}; Petron, Gabrielle^{1,3}; Frost, Gregory J.^{1,2}; Ahmadov, Ravan^{1,2}; Angevine, Wayne^{1,2}; Kim, Si-Wan^{1,2}; Lee, Sang-Hyun^{1,2}; McKeen, Stuart^{1,2}; Trainer, Michael²; Fehsenfeld, Fred¹; Holloway, John^{1,2}; Hsie, Eirh-Yu^{1,2}; Peischl, Jeffrey^{1,2}; Ryerson, Thomas²; Gurney, Kevin⁴

1. CIRES, CU, Boulder, CO, USA
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3. Global Monitoring Division, NOAA, Boulder, CO, USA
4. SOLS, ASU, Tempe, AZ, USA

We developed a new inversion method to calculate an emission inventory for an anthropogenic pollutant without a prior emission estimate at mesoscale. This method employs slopes between mixing ratio enhancements of a given pollutant (CO₂, for instance) with other co-emitted tracers in conjunction with the emission inventories of those tracers (CO, NO_y, and SO₂ are used in this example). The current application of this method employs in-situ measurements onboard the NOAA WP-3 research aircraft during the 2006 Texas Air Quality Study (TexAQS 2006). We use 3 different Lagrangian transport models to estimate the uncertainties introduced by the transport models in the inversion. We demonstrate the validity of this technique by calculating a 4x4 km² emission inventory of anthropogenic CO₂ in the Houston, Texas, area and comparing it to the 10x10 km² Vulcan emission inventory for the United States. The posterior daytime CO₂ inventory for the Houston Ship Channel, home to numerous major industrial and port emission sources, shows excellent agreement with Vulcan, suggesting Vulcan should increase $3\% \pm 27\%$. Compared to Vulcan, the daytime posterior urban CO₂ emissions are higher by between 7% and 20% $\pm 20\%$, depending on the estimates used for net vegetation uptake and ecosystem respiration in Houston. Those differences can be explained by uncertainties in emission factors in Vulcan and by increased emissions from point sources and on-road emitters between 2002, the reference year in Vulcan, and 2006, the year that the TexAQS observations were made.



(a) Map showing the regions of Houston used throughout this paper. (b) NDVI index from MODIS valid in September 2006. (c) CO₂ flux constructed by using 3 tracers in the flux ratio inversion method, used as a prior to calculate (d) a CO₂ posterior in a classic inversion method. (e) Average surface flux of CO₂ in Vulcan and in the CO₂ posterior in the different regions of Houston. (f) Anthropogenic CO₂ emissions in the Vulcan inventory.

Brunner, Dominik

Regional methane sources observed from a motor glider and simulated with a Lagrangian particle dispersion model at kilometre scale resolution

Brunner, Dominik¹; Hiller, Rebecca²; Neininger, Bruno⁴; Künzle, Thomas³; Eugster, Werner²; Nina, Buchmann²

1. Air Pollution/Environ. Technol, Empa, Duebendorf, Switzerland
2. Institute of Agricultural Sciences, ETH Zurich, Zurich, Switzerland
3. Meteotest, Bern, Switzerland
4. Metair AG, Hausen am Albis, Switzerland

According to the Swiss national greenhouse gas inventory, anthropogenic methane (CH₄) sources in Switzerland are strongly dominated by agriculture (83%) followed by waste treatment (10%), fugitive emissions mainly from gas pipelines (4.5%) and other sources such as biomass and fossil fuel combustion (2.5%). Natural sources include emissions from lakes and wetlands but these are estimated to be small. Since most of these emissions are related to microbiological processes with highly variable emission rates depending on environmental factors, CH₄ emission inventories are associated with considerable uncertainties. In order to verify the Swiss inventory and to connect local CH₄ flux measurements over lakes, agricultural land and wetlands with aggregated regional CH₄ fluxes, a series of 17 short aircraft measurement campaigns with a total of 23 flights was performed over

Switzerland in the framework of the project MAIOLICA. CH₄ was measured continuously by a Los Gatos fast methane analyzer mounted in one of the underwing pods of the METAIR-DIMO motor glider. To trace back the potential origin of the measured CH₄ concentrations, Lagrangian backward simulations were started every 3 minutes along the flight tracks using a newly developed version of FLEXPART coupled to the mesoscale numerical weather forecast model COSMO. Model simulated time series of CH₄ concentrations were then obtained by combining the transport simulations with a high-resolution CH₄ emission inventory recently established for Switzerland. In this presentation, we will compare the model simulated and measured CH₄ concentrations and will present a best estimate of CH₄ emissions from different sources by combining the bottom-up (inventory) and top-down (measurements) information in a Bayesian framework.

Cabello, Maria

Influence of interannual variability of transport on surface based measurements of greenhouse gases

Cabello, Maria^{1, 2}; Vermeulen, Alex T.¹; Orza, Jose Antonio G.²

1. ECN, Air Quality & Climate Change, Petten, Netherlands
2. SCOLAB, Física Aplicada, Universidad Miguel Hernandez, Elche, Spain

On the annual to climatic time scales global processes will dominate the variations in observed concentrations of greenhouse gases. Disentangling the different processes at different timescales requires the use of atmospheric transport models that are sufficiently adequate at the required scales in time and space. In this paper we will use trajectory cluster analysis to derive an estimate for the contribution of transport to the variations in greenhouse gas concentrations, as observed at Cabauw tall tower (4.927 E, 51.971 N, -0.7 m a.s.l.) in the period 1992-2011 (Vermeulen et al., 2011). The anomalies in the methane concentration growth rate in this period can at least partly be explained by the changes in global circulation connected to the North Atlantic Oscillation (NAO). The influence of NAO extends to transport directions, mean wind speeds and atmospheric stabilities (Hurrell, 1995). The influence of the use of different meteorological input datasets for the trajectory calculations is also investigated and evaluated using the match between measured and predicted forward concentrations of methane and carbon dioxide at Cabauw and Mace Head station. Winter and summer time high concentrations coincide with low NAO index values while the low concentration periods are often associated with high NAO indices. For carbon dioxide the pattern is less clear due to the fact that winter time concentrations in the northern hemisphere are mainly governed by emissions while summer concentrations are also influenced by assimilation uptake. This research has been financed by an ESF Exchange Grant within the framework of the 'Tall Tower and Surface Research Network for Verification of Climate Relevant Emissions of Human Origin'. References Hurrell, J.W. (1995).

Decadal trends in the North Atlantic Oscillation: Regional temperature and precipitation. *Science*, 269, 676-679.
Vermeulen, A.T., et al. (2011). Greenhouse gas observations from Cabauw Tall Tower. *Atmospheric Measurement Techniques*, 4 (3), 617-644.

Cain, Michelle

A trajectory ensemble study of chemical and physical processes influencing ozone during transport over the North Atlantic

Cain, Michelle¹; Methven, John²; Highwood, Eleanor J.²

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2. Department of Meteorology, University of Reading, Reading, United Kingdom

Chemical transformation during cases of long-range transport across the North Atlantic is studied. Each case was intercepted by research aircraft several times during transport across the North Atlantic as part of the ITCT-Lagrangian 2004 (International Transport and Chemical Transformation) experiment. A Lagrangian photochemical model (Cambridge Tropospheric Trajectory model of Chemistry and Transport, or CiTTYCAT), including a novel treatment for mixing within a trajectory ensemble, is used to simulate the chemical and physical processes acting on the polluted air masses. The modelled change in ozone during transit over the North Atlantic is in agreement with the Lagrangian observations for all cases, taking ensemble spread and observed variability near interceptions into account. Where robust Lagrangian links have been established, the model reproduces the time-averaged OH concentration inferred from hydrocarbon measurements. The Lagrangian model is used to explore the processes acting on the air masses, revealing that the magnitude of net photochemical loss or gain of ozone is greater than the net physical loss or gain in all cases. The physical tendencies often compete - whilst wet and dry deposition are ozone loss processes, mixing can act either to increase or decrease ozone mixing ratios. In many cases the net physical tendency is therefore small. Nevertheless, the model uncertainties associated with physical processes can be large compared with those arising from changes to the photochemical scheme. Physical processes also influence the ozone mixing ratio indirectly through addition or removal of precursor gases. The agreement with observations is weaker in cases of transport at altitudes below 2km, owing to the greater uncertainty in modelling the surface processes of emissions, dry deposition and the boundary layer composition than in the chemistry.

Carver, Glenn D.

Analysis of Aircraft Measurements of Atmospheric Composition by Eulerian and Lagrangian Models

Carver, Glenn D.¹; Cain, Michelle²; Bauguitte, Stephane³; Woolley, Alan³; Purvis, Ruth^{4, 3}; Jones, Rod L.²; Hewitt, Nick⁵; Pyle, John A.¹

1. Department of Chemistry, National Centre for Atmospheric Science (NCAS), Centre for Atmospheric Science, University of Cambridge, Cambridge, United Kingdom
2. Department of Chemistry, Centre for Atmospheric Science, University of Cambridge, Cambridge, United Kingdom
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5. Lancaster Environment Centre, University of Lancaster, Lancaster, United Kingdom

Aircraft measurements of atmospheric composition are inherently difficult to compare with chemistry simulations in Eulerian (gridded) models because of the spatial and temporal scales involved. Aircraft measurements are available at a far higher spatial resolution than found in typical Eulerian models, both horizontally and often vertically. On the other hand, by using Lagrangian models it is possible to analyse trajectories from the flight at a fine spatial resolution but not so easy to simulate chemistry accurately because of the lack of coverage of particles over the domain of interest and mixing between particles. In this presentation, we use a combination of Eulerian and Lagrangian approaches by making use of the p-TOMCAT Chemical Transport Model running at very high resolution and the UK Meteorological Office's Lagrangian dispersion model NAME to analyse aircraft data. Another source of error is the representation of the meteorology in the model compared to that experienced by the aircraft. We present results from the RONOCO (ROLE of Nighttime chemistry in controlling the Oxidising Capacity of the atmosphere) and the OP3 (Oxidant and Particle Photochemical Processes above a south-east Asian tropical rain forest) field campaigns. During RONOCO, aircraft observations were taken of UK and continental outflow around the UK at night. For the OP3 campaign in Borneo flights were carried out to measure the composition just above the forest canopy and in the free troposphere. The NAME model is used to simulate back trajectories from the flights to identify how the air mass histories relate to the different chemical regimes encountered, and how well p-TOMCAT compares to the observations under each regime. We compare the performance of the p-TOMCAT model in these different chemical regimes and show how the model comparison varies. We comment on the differences between the two approaches where a number of sensitivity experiments are presented to demonstrate the advantages and disadvantages of both approaches.

Cassiani, Massimo

A novel approach to model concentration fluctuations and chemical reactions in a dispersing plume based on Lagrangian particle and particle-grid methods

Cassiani, Massimo¹

1. NILU-Norwegian Institute for Air Research, Kjeller, Norway

The mean concentration value is the most important and widely used statistical measure of concentration for a dispersing substance. However, there are applications, as for example the release of toxic or flammables substances, where the concentration fluctuations, generated by the turbulent nature of the flow, are of importance. In these cases the concentration fluctuations need to be accounted for with the prediction of at least the second concentration moment. Moreover, it is known that concentration fluctuations may influence the reaction rates of a chemically reactive plume with a decrease of the reaction rates for non premixed chemical species, or an increase of the reactions rates for premixed chemical species. These effects are very difficult to capture in a model. Here, initially a novel approach is proposed to predict concentration fluctuations for a non reactive plume in the framework of one particle Lagrangian stochastic models. The model results are compared to the wind tunnel experiments of Fackrell and Robins (Journal of Fluid Mechanics, 117, 1982) for the dispersion from a point source in a neutral boundary layer and a satisfactory agreement with the measurements is shown for both the mean and the variance of the concentration. Subsequently, it is discussed the extension of the proposed approach to model the dispersion of a chemically reactive plume. This extension is based on the use of surplus/deficit concentrations with respect to the background. Results are shown for the simulation of a NO plume emitted in an O₃ background and they are compared with the experimental measurements of Brown and Bilger (Journal of Fluid mechanics, 312, 1996; Atmospheric Environment, 32, 1998). Despite the fact that the influence of turbulence on the chemical reactions rates is accounted for only in an approximated manner, the simulation's results show an encouraging agreement with the experimental measurements. Therefore, the comparison demonstrates the feasibility to model plume chemical reactions with the proposed method. All these characteristics qualify this novel approach as a good candidate to be used for plume-in-grid modeling in mesoscale and large scale chemistry transport models.

Chen, Huilin

Simulations of carbonyl sulfide (COS) and CO₂ for North America using STILT

Chen, Huilin¹; Petron, Gabrielle^{1,2}; Trudeau, Michael^{1,2}; Karion, Anna^{1,2}; Kretschmer, Roberto³; Gerbig, Christoph³; Campbell, Elliott⁴; Nehrkorn, Thomas⁵; Eluszkiewicz, Janusz⁵; Miller, Ben^{1,2}; Montzka, Stephen¹; Jacobson, Andrew R.^{1,2}; Sweeney, Colm^{1,2}; Andrews, Arlyn¹; Tans, Pieter¹

1. NOAA/Earth System Research Laboratory, Boulder, CO, USA
2. Cooperative Institute for Research in Environmental Sciences, Boulder, CO, USA
3. Max Planck Institute for Biogeochemistry, Jena, Germany
4. Sierra Nevada Research Institute, University of California, Merced, CA, USA
5. Atmospheric and Environmental Research, Inc., Lexington, MA, USA

Understanding biospheric CO₂ fluxes is paramount if climate studies are to be able to analyze the response of terrestrial ecosystems to climate change and monitor fossil fuel emissions reductions. Carbonyl sulfide (COS) may be a useful tracer to provide a constraint on photosynthesis [gross primary production (GPP)]. Here we simulate both COS and CO₂ using the Stochastic Time-Inverted Lagrangian Transport (STILT) model coupled with various biospheric fluxes, such as fluxes estimated from the Vegetation Photosynthesis and Respiration Model (VPRM), CarbonTracker, and from the Carnegie-Ames-Stanford Approach (CASA) model. The STILT model is driven by Weather Research and Forecast (WRF) meteorological fields. The WRF-STILT system is compared with the STILT driven by the ECMWF (European Center for Medium range Weather Forecasting) meteorology for the North American domain. For this study, measurements of COS and CO₂ from the NOAA/ESRL tower and aircraft air sampling networks will be used. The biospheric COS fluxes will be estimated from a GPP-based model coupled with the above mentioned biospheric fluxes. Other COS fluxes, such as anthropogenic, biomass burning and soil uptake will be derived based on previously existing analyses of temporal and spatial variations. The lateral boundary conditions for COS will be obtained from a recent global 3-D COS model or the modeled atmospheric concentrations of COS using the Sulfur Transport Eulerian Model (STEM). An empirical curtain derived from observations will be used for CO₂ lateral boundary conditions. Comparison of the simulations for both COS and CO₂ using various biospheric fluxes provides an opportunity to assess the performance of these biospheric models. In addition, we will assess an inversion analysis for both COS and CO₂, in an effort to improve the performance by applying constraints separately on GPP and respiration in the near future.

D'Amours, Réal

Application of Lagrangian modelling to radioactive releases following the Fukushima nuclear accident

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Comprehensive data sets to validate large scale transport and dispersion models are rather few. The European Tracer Experiment (ETEX) was held in 1994, seventeen years ago, and produced two data sets. Understanding the performance of dispersion models with regard to the second data set is still a challenge (Potemski et al. 2008). Accidental releases of radioactive substances in the atmosphere may provide opportunities to validate models, because these substances can be detected at very low concentrations and are closely monitored worldwide. Radioactive substances released from the Fukushima nuclear power plant, following the devastating tsunami of March 11 2011, were measured at many locations over the Northern Hemisphere. During the initial phases of the accident, the Canadian Meteorological Centre used its Lagrangian Dispersion Models to provide advice on the fate of the radioactive plume in fulfilment of its national and international commitments. In this paper, results of forward and inverse simulations are compared to measurements of radioactive tracers from Japan and North America. Estimates of released quantities based on the observations and inverse modelling are also assessed in light of the available information. An evaluation of different modelled processes (and possible deficiencies) is also attempted by comparing results for aerosols and noble gases. The paper focuses on the early stages of the accident.

Dacre, Helen

Evaluating the structure and magnitude of the ash plume during the initial phase of the 2010 Eyjafjallajökull eruption using lidar observations and NAME simulations (*INVITED*)

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The Eyjafjallajökull volcano in Iceland erupted explosively on 14 April 2010 emitting a plume of ash into the atmosphere. The ash was transported from Iceland towards Europe where mostly cloud-free skies allowed ground-based lidars at Chilbolton in England and Leipzig in Germany to estimate the mass concentration in the ash cloud as it passed overhead. The UK Met Office's Numerical Atmospheric-dispersion Modelling Environment, NAME, has been used to simulate the evolution of the ash cloud from the Eyjafjallajökull volcano during the initial phase of the ash emissions, 14-16 April 2010. NAME captures the timing and

sloped structure of the ash layer observed over Leipzig, close to the central axis of the ash cloud. Relatively small errors in the ash cloud position, probably caused by the cumulative effect of errors in the driving meteorology en route, result in a timing error at distances far from the central axis of the ash cloud. Taking the timing error into account, NAME is able to capture the sloped ash layer over the UK.

Comparison of the lidar observations and NAME simulations has allowed an estimation of the plume height time-series to be made. It is necessary to include in the model input the large variations in plume height in order to accurately predict the ash cloud structure at long range. Quantitative comparison with the mass concentrations at Leipzig and Chilbolton suggest that around 3% of the total emitted mass is transported as far as these sites by small (< 100 micrometer diameter) ash particles.

Denlinger, Roger P.

Objective and quantitative comparison of volcanic ash dispersal from different transport models

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Volcanic eruptions often spew fine ash tens of kilometers into the atmosphere. This ash is then carried downwind, disrupting air traffic and posing a hazard to air travel. To mitigate such hazards, the community studying ash hazards must assess risk of ash ingestion for any flight path and provide robust and accurate forecasts of volcanic ash dispersal. A number of different transport models have been developed for this purpose. To evaluate ash dispersal estimates from a given model as well as compare estimates among different models, we provide two solutions: 1) a framework for optimal forecasts and their uncertainties given any model and any observational data, and 2) a quantitative and objective method to compare results from different transport models. The first solution involves random sampling of the probability distributions of input (source) parameters to a transport model, and running the model iteratively with different inputs, each time assessing the predictions that the model makes about ash dispersal. These predictions are embodied in a likelihood function based on a comparison of model output to observational data (usually an ash cloud imaged by satellite remote sensing). The minimum misfit between model output and observations determines the maximum likelihood estimate, which is the volcano source input that provides the best fit to observations. Model comparison in (2) is a more difficult task, because with successive refinement more complex models can always be built to maximize likelihood. Bayes theorem provides a way out of this conundrum, as it penalizes models for increased complexity without constraint. To compare the results of two different transport models in (2), we determine maximum likelihoods as in (1) for each model. We then use Bayes theorem to combine the maximum likelihood estimation of each model with a data-dependent measure of the sensitivity of model output to

input data to define the evidence for that model. We have applied this method to recent eruptions of Redoubt volcano in Alaska and Eyjafjallajökull in Iceland to compare of the evidence for an Eulerian model (Ash3d) and a Lagrangian model (PUFF) and thereby illustrate the strengths and weaknesses of each approach. This comparison of evidence benefits from the physical characteristics of ash dispersal from an eruption column. Volcanic ash clouds typically form ribbons of ash much longer than they are wide. In model comparisons, the sharp boundaries of ash clouds constrain well-defined maximum likelihood functions, even in complex wind fields that create multiple ribbons' of ash. Accurate satellite detection of these ribbon-like ash clouds then allows us to successively refine our judgments as to the efficacy of each model's ability to simulate ash dispersal.

Devenish, Benjamin

The geometry of turbulent dispersion

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The dispersion of a pair of marked particles is a celebrated problem in the study of turbulence and is ideally suited to the Lagrangian framework. Its importance is both because of its connection with concentration fluctuations and the insight it provides into the structure of turbulence. With the aid of three Lagrangian stochastic models (LSM), two quasi-one-dimensional models with and without intermittency effects and a three-dimensional model with Gaussian distributed relative velocity statistics, I will show that the pairs which separate slowly play an important role in the separation process. Since C_0 , the constant of proportionality in the second-order Lagrangian structure function, is prescribed in the LSM, it is possible, by varying the value of C_0 , to assess and quantify the relative importance of ballistically and diffusively separating pairs. I will show that even for values of C_0 between 4 and 6, the values typically associated with real turbulence, diffusively separating pairs strongly influence the separation statistics (at least at low order). Furthermore, the differences between the three models both with each other and with direct numerical simulation (DNS) of turbulence highlight the importance of rotation in estimating the correct rate of separation. In recent years, the dispersion of four marked particles or tetrads in 3-D turbulence has been studied both experimentally and in DNS providing more detailed information on the geometry of turbulence. In particular, turbulence tends to flatten or elongate the tetrads. The analysis of Lagrangian four-point velocity-difference statistics in DNS shows that inertial-range eddies typically generate a straining field with a strong extensional component aligned with the direction of elongation and weak extensional/compressional components in the orthogonal plane. A recently developed LSM for the dispersion of tetrads with Gaussian velocity statistics reproduces many of the features of tetrad dispersion in DNS. In particular, the model also tends to produce elongated shapes with a local velocity field that has two extensional components, one strong and one weak and one

compressional component. I will present various statistics from the new LSM and discuss the effect of varying C0 on the shape and size statistics of the tetrads.

Draxler, Roland

Data Archive of Tracer Experiments and Meteorology (DATEM)

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A common theme for dispersion model users is the need to understand the effects of changing model parameters and meteorological data. A first step was accomplished some time ago by standardizing the format of several tracer experiments in which NOAA participated, providing meteorological data (the 2.5 degree global reanalysis), and statistical analysis software. The DATEM project initially provided only anonymous FTP access. The six experiments currently available include a wide range of durations and distances: from the urban scale Metropolitan Tracer Experiment to the continental scale Across North America Tracer Experiment. The DATEM interface (http://ready.arl.noaa.gov/HYSPLIT_datem.php) has been updated to permit anyone to interactively run the NOAA dispersion model (<http://ready.arl.noaa.gov/HYSPLIT.php>) for any of the experiments. With the availability of the 32-km North American Regional Reanalysis, dispersion simulations become more realistic. As part of the web evaluation, a common set of statistical analyses are performed on the results of each simulation. Users can determine the sensitivity of changing model parameters on the accuracy of the calculation. After selecting an experiment, several menu options appear in sequence, such as which meteorological data file to use, the dispersion equations (3D particle or puff), and the turbulence parameterizations (how the mixing is computed from the meteorological data fields). When the simulation completes, the statistical results are available in tabular form, as well as a simple graphic showing how the measured-calculated scatter diagram of the current simulation compares with a base simulation. The web interface effectively provides a self-directed tutorial in understanding the importance of various model parameterizations and the significance of the meteorological data. The accuracy of a plume dispersion calculation is primarily a function of how well the gridded meteorological data, discrete in space and time, represent the continuous fields of winds and temperature. The next phase of the DATEM project will provide finer spatial and temporal resolution meteorological data for each experiment. Several issues need to be resolved to determine the optimal output resolution. The output data volume can be huge and finer spatial resolution also requires finer temporal resolution. WRF output fields are usually for a specific time. Time averaging the output may mitigate the temporal resolution requirements. Tests using the base 27-km WRF fields for CAPTEX release #2 showed substantially

better model performance, in terms of a higher correlation (0.74 | 0.59), lower NMSE (10.4 | 26.7) and fractional bias (-0.03 | 0.37), and greater overlap of predicted and observed plume (73% | 63%), than the NARR meteorology. Using even finer spatial resolution (9, 3, and 1 km) had no influence on the results. Simulations with data assimilation showed a degraded performance compared with the simulation without observational data nudging. Additional tests are being conducted with higher temporal resolution output data. After a WRF configuration is selected, meteorological data fields will be created for all experiments and be made available through the DATEM web page.

Duran Quesada, Ana Maria

Role of the Caribbean Low Level Jet in the transport of moisture in the Intra Americas Sea region: A Lagrangian study based on backward trajectories

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Due to the importance of the Caribbean Low Level Jet (CLLJ) study on its role for moisture transport processes in the Intra Americas Sea (IAS) was proposed. A set of backward trajectories generated with the Lagrangian dispersion model FLEXPART for the 1980-1999 period is used to identify the trajectories associated with the CLLJ in a climatological basis. Two clustering methods were applied to the data, based on the Dorling method a first set of clusters was obtained. A second clustering based on the dynamic features of the CLLJ was applied to obtain those mean trajectories attributable to the CLLJ. It was determined that the method does not allow just the identification of the CLLJ but also its more important features regarding the mean seasonal position of the structure and moisture content. The seasonal cycle of the CLLJ was properly represented, showing its importance during boreal winter in the transport of moisture to Central America as well as the development of its northward branch during summer. The importance of the CLLJ during summer was studied fundamentally considering the transport of moisture from the Caribbean Sea to the Gulf of Mexico and further contributions to the North American Monsoon System (NAMS). Beyond the analysis of the seasonal cycle, the interannual variability was also analysed. As expected, the variability of the trajectories presented a response to El Niño-Southern Oscillation (ENSO) forcing. Some evidence on the importance of the forcing of the NAO is in agreement with the hypothesis of the role of the NASH modifying the CLLJ. For both seasonal and interannual analysis, the relation between the contribution of the transport of moisture through the CLLJ and precipitation in Central America was studied. A coherent pattern between the seasonal cycle of the intensity of the CLLJ and precipitation was found. Moreover, the response of precipitation in Central America to ENSO was in phase with determined

patterns presented by the CLLJ trajectories during the same ENSO events. The applied methodology allowed the identification of the trajectories associated to the CLLJ, the study of the main features of the variability of the CLLJ and its role in the transport of moisture associated with precipitation in the surrounding regions. Finally, based on the results, further questions that may be of scientific interest are proposed.

Duran Quesada, Ana Maria

Variability of sources of moisture in the Intra Americas Sea region based on a Lagrangian approach

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A set of backward Lagrangian trajectories was computed using the Lagrangian model FLEXPART with ERA-40 Reanalysis data as input for the twenty years period 1980-1999. The climatological main moisture sources for regions of interest in the Intra Americas Sea (IAS) region named Central America and southwestern North America were identified. In agreement with previous results, the surrounding oceanic region was determined to be the main source of moisture for Central America. Furthermore, the results have confirmed the result that in the case of the development of the North American Monsoon the contributions of moisture come from both ocean and continental region. The method allowed to track the air masses from different locations to the NAM region to determine the importance of the contributions from each source. The tracking of air masses enabled the reproduction of the variations in the contribution from the sources during the development of the NAM. This, showing how the importance of the sources varies in function on the development phase of the monsoon. Variability associated with ENSO (El Niño- Southern Oscillation) and WHWP (Western Hemisphere Warm Pool) was also studied. Regarding Central America, the results showed a good agreement between the variations in the magnitude of the sources of the sources of moisture and significant modifications in the regional precipitation patterns. The occurrence of severe droughts in western Central America was determined to be partially caused by a deficit in the contribution of moisture from regional oceanic sources. An extended analysis on the severe precipitation events in Central America associated with ENSO was performed in order to determine whether or not these events were attributable to the variability of the oceanic sources of moisture.

Eckhardt, Sabine

The impact of North American and Asian emissions on carbon monoxide and ozone concentrations over Europe

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While there exist many model and case studies of intercontinental transport, to date no statistical analyses of measurement data over Europe exist, which analyze the influence of North American and Asian emission sources. In this study, we used a large data set of carbon monoxide (CO) and ozone measurements to examine the influence of American and Asian emissions on the chemical composition of the atmosphere over Europe. We analyzed 13 years (1996-2009) of ozone and 7 years of CO measurements from the MOZAIC programme taken during ascent and descent from European airports of commercial airliners equipped with instruments measuring meteorological parameters as well as some trace gases. We determined the source regions influencing the measurements with the Lagrangian particle dispersion model FLEXPART. CO and ozone data were averaged over 1 km high layers during ascent and descent and for each individual 1-km-averaged measurement, 40000 particles were released and followed 20 days backward in time. For the entire period, we performed backward calculations for 160000 measurements. Using the EDGAR emission inventory and the FLEXPART backward calculation, CO concentration as predicted by the model could be derived. Comparing the modeled and simulated CO values shows that measured CO and modeled CO enhancement are well correlated and, thus, that the model captures the relevant transport processes. Subsequently, we grouped the MOZAIC measurements according to the dominant source regions, distinguishing between European and North American-dominated measurements as well as Asian-dominated measurements, respectively. A seasonal analysis shows that there are clear differences in the ozone to CO ratio depending on the source region. The importance of the North American emissions for Europe shows a weak seasonal variation. Altitudes and regions which receive the highest pollution load from intercontinental transport are identified.

Eluszkiewicz, Janusz

Lagrangian Modeling of Satellite Greenhouse Gas Observations

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Quantification of greenhouse gas (GHG) fluxes at regional and local scales is required by the Kyoto protocol and potential follow-up agreements, and their accompanying

implementation mechanisms (e.g., cap-and-trade schemes and treaty verification protocols). Dedicated satellite observations, such as those provided by the Greenhouse gases Observing Satellite (GOSAT), the upcoming Orbiting Carbon Observatory (OCO-2), and future active missions, e.g., Active Sensing of CO₂ Emissions over Nights, Days, and Seasons (ASCENDS) and Advanced Space Carbon and Climate Observation of Planet Earth (A-SCOPE), are poised to play a central role in this endeavor, complemented by observations “of opportunity” afforded by existing instruments (particularly SCIAMACHY, TES, AIRS, and IASI). In order to prepare for future dedicated GHG monitoring missions and quantify the ability of current satellite observations to constrain GHG fluxes at high spatial and temporal resolution, we are applying the Stochastic Time-Inverted Lagrangian Transport (STILT) model driven by meteorological fields from a customized version of the Weather Research and Forecasting (WRF) model to generate surface influence functions for satellite GHG observations. These “footprints” (or adjoint) express the sensitivity of observations to surface fluxes in the upwind source regions and thus enable the computation of a posteriori flux error reductions resulting from the inclusion of satellite observations (taking into account the vertical sensitivity and error characteristics of the latter). Several features make WRF-STILT an attractive tool for regional analysis of satellite observations: 1) WRF meteorology is available at higher resolution than for global models and is thus more realistic, 2) The Lagrangian approach minimizes numerical diffusion present in Eulerian models, 3) The WRF-STILT coupling has been specifically designed to achieve good mass conservation characteristics, and 4) The receptor-oriented approach offers a relatively straightforward way to compute the adjoint of the transport model. These aspects allow the model to compute surface influences for satellite observations at high spatiotemporal resolution and to generate realistic flux error and flux estimates at policy-relevant scales. The main drawbacks of the Lagrangian approach to satellite simulations are inefficiency and storage requirements, but these obstacles can be alleviated by taking advantage of modern computing resources (e.g., AER has been running WRF-STILT on the NASA Pleiades supercomputer as part of analysis and design studies for TES and ASCENDS). This research builds upon and extends other efforts to develop and apply the WRF-STILT model to regional top-down GHG flux estimates, with examples to be given in this presentation and the companion paper by Nehr Korn et al. We gratefully acknowledge funding by NASA, NSF, and NOAA.

Esler, James G.

A unified framework for global Eulerian and Lagrangian models of atmospheric transport

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Is it possible, and / or desirable, to develop a theoretical and computational framework that permits an exact correspondence between Eulerian and stochastic Lagrangian models of atmospheric transport? The basic idea behind such a framework is, given a particular Eulerian evolution equation (which may include representations of convection and boundary layer turbulence), to select a Markov process for the corresponding Lagrangian trajectory model that has a formally identical master equation (Fokker-Planck equation) for the evolution of its probability distribution. The advantages of the proposed framework are both theoretical, as each type of model can be regarded as discretizations of the same underlying set of equations, and practical in that results from each type of model can be used for accurate cross-validation. Monte-Carlo simulations using the Lagrangian model can even be used to estimate the numerical diffusivities of different Eulerian advection schemes. Each of the above points is illustrated using models of a simple advection-diffusion problem. Some of the difficulties and potential pitfalls encountered in adapting global Eulerian and stochastic Lagrangian transport models to the proposed framework are discussed, such as mass conservation, implementation of different convection and boundary layer schemes, and accounting for the implicit diffusivity of Eulerian advection schemes. Finally some preliminary comparisons from global models, namely the Eulerian model TOMCAT and the Lagrangian model FLEXPART, are discussed.

Ferrero, Enrico

Beyond the Limits of the Lagrangian Particle Models, From the Chemistry to the Entrainment

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It is well known that the Lagrangian particle models, even though satisfactorily used to simulate the dispersion in many complex situations, encompass some limits in dealing with the interaction of the plume with the background. This can be either the ambient air or substances mixed with it, such as reactive pollutants. The limitation is substantially due to the huge amount of particles that are necessary to fill the computational domain. Here we propose a method to overcome this limitation allowing to simulate, for example, the chemical reactions of a NO plume with the background ozone or the air entrainment during plume rise. The method is based on the concept of “deficit”, i.e. a fictitious new

scalar carried by each particle. The deficit is the difference between the background species concentration carried / transported by the plume and its background value. This allows to evaluate the in-plume variations, due to chemical or physical processes, of the species without considering particles that do not belong to the plume, sparing considerable computation time. Some applications of the deficit method to different dispersion cases are presented and discussed.

Fleming, Zoë

Untangling the seasonal and geographical influences at Cape Verde Atmospheric Observatory

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The Met office's NAME atmospheric dispersion model has been used to track the air arriving at the Cape Verde Observatory in the Atlantic Ocean. The Observatory receives air masses that originate from north America, Europe, Saharan Africa as well as air masses that have not passed over a land mass for a number of days and are representative of clean Atlantic or marine coastal air. 3 hourly footprints of the air mass pathway over the previous 10 days were used to construct a library of regional influences which have been classified into seven main air mass types influencing the station. The Cape Verde Observatory has been measuring a variety of trace gases since October 2006 and is a Global WMO-GAW station and is run as a collaboration between research institutes in the UK, Germany, the US and Cape Verde. The dataset has been analysed in combination with dispersion modelling to investigate variations in trace gases with air mass history over the past 5 years. The aim is to detect seasonal and regional variations in atmospheric composition with the intent of identifying intra-annual and eventually long term trends according to air mass history. This will help us to understand source-receptor relationships and the role of long range transport to marine boundary layer sites.

Franzese, Pasquale

PDF Modeling of the Internal Fluctuations of a Tracer Release

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A well-established research approach to the calculation of concentration fluctuations at fixed points in space is based on the modeling of the internal concentration field, namely the concentration statistics measured in the frame of reference relative to the center of mass of the cloud, coupled to the statistics of its centerline position, as originally proposed by Gifford (1959). Following this idea, analytical Gaussian plume models, and Lagrangian stochastic models for fluctuating plumes have been developed. However, these models are not capable of calculating the required relative concentration statistics, which are thus parametrized. The random transverse transport of the internal scalar fluctuations can be used to represent the scalar fluctuations at a fixed point downstream of the source. In addition, if the release is instantaneous, statistics at fixed distances from the center of mass are more representative than statistics at fixed points in space. We present a probability density function (PDF) model for the statistics and the probability of concentration in the frame of reference relative to the center of mass of the cloud. Turbulent dispersion in the relative frame of reference in one dimension is represented by a system of four Lagrangian stochastic equations. Micromixing is represented by an interaction by exchange with the conditional mean (IECM) model modified to be applied to internal statistics. We show results for the transverse profiles of the first four moments of concentration, and for the full concentration PDF at different longitudinal distances from the source and at different transverse distances from the centre of mass. Field and laboratory measurements are also reported for comparison.

Galmarini, Stefano

Multi-model model ensemble dispersion modelling: from empiricism to structure (*INVITED*)

Galmarini, Stefano¹

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Over the past decade the Joint Research Center has been engaged in the study of ensembles of atmospheric dispersion and air quality models. Starting from long range dispersion models to three-dimensional eulerian air quality models, a large variety of data sets have been analyzed in many different ways in a first attempt to try to improve the quality. The acquired experience has led to the identification of the weaknesses and advantages of the technique to the extent that a rational framework could be identified that went beyond the pure practices of putting model results and hoping that the ensemble was better. The paper aims at presenting the outcomes of this research and to identify the

many still opened questions that need to be addressed. Several are in fact the pitfalls that can be encountered when using this technique that especially in the case dispersion forecasting may well lead to surprises.

Gerbig, Christoph

Assessment of Network design for greenhouse gas observing systems using Lagrangian footprint simulations

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Deriving greenhouse gas balances at regional scales can be done via two approaches: up-scaling of local information such as from inventories for emissions and natural processes (“bottom-up”), or independently via combined atmospheric observations and inverse transport modeling (“top-down”). For the top-down approach, a dense coverage of the atmosphere with an observing network that enables a precise estimation of surface fluxes is associated with considerable costs. To facilitate decision-making with respect to the number and location of observing stations, a quantitative network design tool is required that allows balancing cost and benefit. Here we assess different aspects of network design using the Stochastic Time Inverted Lagrangian Transport model. We present sensitivities of mixing ratio observations from various potential networks to surface-atmosphere exchange fluxes, assess impact from transport model uncertainties, and investigate the impact of different a-priori assumptions for flux distributions on the potential to retrieve fluxes at reduced uncertainty bounds.

<http://www.bgc-jena.mpg.de/~christoph.gerbig>

Guerrero, Oscar J.

Assessing wetland and anthropogenic methane fluxes in Colombia and Panama from Lagrangian simulation analysis of aircraft-borne measurements during TC4

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Methane is the second most important long lived greenhouse gas (GHG) in the Earth’s atmosphere accounting for ~20% of the positive radiative forcing. The first step towards developing GHG mitigation strategies is to obtain sufficiently accurate and detailed source and sinks estimations. While ~2/3 of the global methane emissions are anthropogenic, the wetlands are the single largest source. Therefore, in many cases, wetland emissions must be included in inverse modelling calculations aimed at validating anthropogenic emission inventories from ambient air concentration measurements. High accuracy and precision CH₄ measurements over Colombia and Panama carried out in 2007 during NASA’s TC4 mission revealed elevated methane enhancements (up to ~500 ppbv over Uraba, Colombia). We used STILT to estimate methane mixing ratios based on surface fluxes at regional level over four regions of both Colombia and Panama. STILT was applied along with assimilated meteorological fields from GDAS and ECMWF and a priori methane inventories for anthropogenic (EDGAR) and wetland emissions (Kaplan’s and Matthews and Fung’s). The modeled mixing ratios were compared to the TC4 mission measurements. We calculated errors in meteorological fields by comparison with meteorological observations and propagated them into the CH₄ mixing ratio uncertainties. A Bayesian inversion analysis was performed to assess the effects of transport errors on modeled mixing ratios and to adjust the a priori fluxes. We obtained scaling factors for all the selected receptors over the whole domain of study and for each one of the four regions. In general, the Kaplan’s wetlands and anthropogenic fluxes were adjusted by scaling factors of 0.6 and 1.0, respectively. Although the a posteriori enhancements show a better agreement with measurements, we found that the discrepancies cannot be reduced for the 4 regions simultaneously. This fact points to meteorology as the main source of error. Similar results were obtained using the Matthews and Fung’s wetlands inventory. This is not

surprising bearing in mind the complexity of horizontal and vertical transport in the Intertropical Convergence Zone, particularly over the strong moist convection period experienced during TC4.

Hansen, Ayoe B.

Test of Semi-Lagrangian Methods for Advection in the Danish Eulerian Hemispheric Model

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For air pollution modeling with high resolution, advection schemes of high accuracy and efficiency are essential. In general the advantages of replacing a traditional Eulerian advection scheme with a semi-Lagrangian method can be numerous. The purpose of the present study is to develop a model covering a limited area, specifically Denmark, with very high resolution, in this case 1 km by 1 km. The increase in spatial and there by temporal resolution must in part be compensated by a more efficient method in order for the model to be applicable to forecasting and the like. In Hansen et al. (2010) several semi-Lagrangian methods were developed and tested on idealized two dimensional tests cases, the traditional slotted cylinder, see e.g. Zerroukat et al. (2002), or the rotating cone, see e.g. Molenkamp (1968) and Crowley (1968), with chemistry and compared to the Eulerian advection scheme, Accurate Space Derivatives. A classical 3D cubic semi-Lagrangian advection scheme is implemented and tested in the Danish Eulerian Hemispheric Model (DEHM). As for the idealized test cases, the focus is accuracy, local mass conservation, and computational efficiency, in respect to high resolution air pollution modeling over a limited domain. The advantage of semi-Lagrangian methods over strictly Eulerian methods, i.e. the possibility of long timestepping without development of instability versus the ability of the advection scheme used in DEHM, the Accurate Space Derivatives, to model peak concentrations in the idealized test cases are investigated in order to find the most accurate and efficient scheme for very high resolution over a limited area. The present study is a part of the research of the Center for Energy, Environment and Health, financed by The Danish Strategic Research Program on Sustainable Energy. References Crowley, W. P., January 1968. Numerical Advection Experiments. *Monthly Weather Review* 96 (1), 1–11. Hansen, A. B., Brandt, J., Christensen, J. H., Kaas, E., 2010. Semi-lagrangian methods in air pollution models. *Geoscientific Model Development Discussions* 3 (4), 2361–2438. URL <http://www.geosci-model-dev-discuss.net/3/2361/2010/> Molenkamp, C. R., April 1968. Accuracy of Finite-Difference Methods Applied to the Advection Equation. *Journal of Applied Meteorology* 7, 160– 167. Zerroukat, M., Wood, N., Staniforth, A., 2002. SLICE: A Semi-Lagrangian Inherently Conserving and

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Haynes, Peter

The advection-condensation paradigm for stratospheric water vapour

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Lower stratospheric water vapour has important implications for the radiative balance of the troposphere and for stratospheric ozone chemistry. A key question has been ‘What processes determine stratospheric water vapour concentrations and their seasonal and interannual variations’. The advection-condensation (A-C) paradigm, which postulates that water vapor concentrations are governed to leading order by Lagrangian transport through a temperature (and hence saturation mixing ratio) field that varies in both space and time has been shown to provide a useful leading-order description. Historically it has seemed difficult to explain low stratospheric water vapour concentrations on the basis of saturation mixing ratios calculated from observed large-scale temperatures. However Lagrangian calculations such as those of Fueglistaler et al (2005) showed that provided the sampling of the space and time variations of the large-scale temperature field were taken into account there was good agreement between predicted and observed water vapour concentrations. More recently (Liu et al, 2010) the Lagrangian calculations have been refined to use trajectories based on improved meteorological analyses, in some cases using diabatic heating information to calculate vertical velocity. These calculations seem to predict water vapour concentrations that are anomalously low. Careful analysis, reported in Liu et al (2010), establishes that the anomaly is significant relative to the uncertainty in the calculation and therefore makes it worthwhile to consider what extra physics might be added to the A-C paradigm to increase predicted water vapour concentrations to bring them closer to those observed. Fueglistaler, S., Bonazzola, M., Haynes, P.H., Peter, T., 2005: Stratospheric water vapor predicted from the Lagrangian temperature history of air entering the stratosphere in the tropics *J. Geophys. Res.*, 110, D10, D10S16, doi:10.1029/2004JD005516. Liu, Y.-L., Fueglistaler, S., Haynes, P. H., 2010: The advection-condensation paradigm for stratospheric water vapour, *J. Geophys. Res.*, 115, D24307, doi:10.1029/2010JD014352.

Heard, Imogen P.

A Comparison of Atmospheric Dispersion Model Predictions with Observations of SO₂ and Sulphate Aerosol from Volcanic Eruptions

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The Lagrangian Numerical Atmospheric-dispersion Modelling Environment (NAME) has previously been used operationally to model volcanic ash at the London VAAC (Volcanic Ash Advisory Centre), including that from the eruptions in Iceland of Eyjafjallajökull in 2010 and Grímsvötn in 2011. To prepare the model for possible future eruptions, the ability of NAME to model the release and dispersion of volcanic SO₂, the chemical processes leading to the production of sulphate aerosol, and the subsequent dispersion of sulphate aerosol, has been investigated. The eruptions of Sarychev in 2009, Kasatochi in 2008 and Eyjafjallajökull in 2010 were simulated and results for SO₂ and sulphate aerosol optical depth (AOD) were compared with satellite observations. NAME results compare favourably with available observations in terms of both geographical distribution and air concentration for all three cases. NAME modelled concentrations of SO₂ are of the same order of magnitude as those observed by satellite for the eruption of Sarychev, and over 70% of modelled values of sulphate AOD are within a factor of 2 of those observed for both Sarychev and Kasatochi. Although significant uncertainties are present in both the model and observations, this work shows that NAME is able to model SO₂ and sulphate from volcanoes. Therefore NAME could be used for this purpose, in addition to volcanic ash modelling, in future eruptions where significant quantities of SO₂ are released.

Henne, Stephan

Deposition of trifluoroacetic acid (TFA) resulting from future emissions of HFO-1234yf from mobile air conditioners in Europe

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The cooling agent HFC-134a (1,1,1,2-Tetrafluoroethane), widely used in today's mobile air conditioners, has a high 100-year global warming potential (GWP) of 1430 and therefore will be phased out. The hydrofluoro-olefin HFO-1234yf (2,3,3,3-tetrafluoropropene) is a promising replacement with a low 100-yr GWP of 4. The only final degradation product of HFO-1234yf in the atmosphere is

trifluoroacetic acid (TFA), which is formed by initial reaction of HFO-1234yf with OH or Cl radicals, followed by rapid hydrolysis on cloud droplets. TFA is weakly phytotoxic and persistent in the environment, therefore it might pose a risk for aquatic biomes. Average atmospheric HFO-1234yf lifetimes with respect to OH solely are about 10 to 12 days and about 6 days if Cl is considered as well. Thus, HFO-1234yf is expected to degrade relatively quickly and not undergo significant hemispheric scale transport. As a result, the expected deposition of TFA might occur relatively concentrated and close to the emission sources. We analyzed TFA deposition, resulting from emissions of HFO-1234yf after a complete conversion of the European vehicle fleet, with two Lagrangian transport models. Approach (A) utilized the Lagrangian Particle Dispersion Model FLEXPART with a detailed atmospheric transport description but prescribed OH and Cl concentration fields. Approach (B) was based on the Lagrangian chemistry transport model CRI-STOCHEM that includes a tropospheric chemistry scheme, but a less detailed transport description. Both models used a low and high emission scenario as input, with total European emissions of approximately 10 and 20 Gg of HFO-1234yf per year, respectively. In approach (A) annual mean mixing ratios of HFO-1234yf were between 1.5 and 2.6 ppt (low and high scenario) in Europe with hot spots in Northern Italy and the Ruhr area (Germany). In approach (B) annual mean mixing ratios of 0.6 – 1.3 ppt were estimated. Mean mixing ratios of TFA over Europe were 0.05 – 0.11 ppt in both approaches. Largest mixing ratios were simulated for Northern Italy and the eastern Mediterranean. This resulted in total (dry + wet) annual average deposition of 0.37 – 0.65 kg km⁻² yr⁻¹ (A) and 0.41 – 0.80 kg km⁻² yr⁻¹ (B). The highest deposition rates were estimated for the summer months and over northern Italy, southern Germany, and the Mediterranean. Annual average TFA concentrations in rainwater in Europe were 0.3 – 0.6 µg L⁻¹ (A) and 0.4 – 0.8 µg L⁻¹ (B). Maxima were observed over southern Germany and the Czech Republic. Individual seasonal averages indicated greater concentrations in summer and maxima over the continent were in the range of 3 – 5 µg L⁻¹. These predicted seasonal maximal TFA rainwater concentrations were at least one order of magnitude smaller than the no-effect level of 120 µg L⁻¹ that was estimated in toxicological studies for the most sensitive algae. However, it is advisable to monitor TFA deposition and rainwater concentrations in the future and to study its accumulation in sensitive biomes in arid areas.

Hernández, Jesús A.

EDA s Classifier Systems for Identifying Patterns of Particulate Matter (PM_x) Dispersion in a Study Area

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One of the main questions when it comes to mitigating the effects of particulate matter PM₁₀ concentration leads to an area of study is the fact to determine their behavior in space over time. That is why in this article, we present a classifier based on the principles of the estimating distribution algorithms EDAs (Hernández & Opsina, 2010), to identify patterns hours wind fields which determine the dynamics of dispersion PM₁₀ particulate matter in an area of study (Peña P., Hernández R., & Toro G., 2010(a)). The first instance, this model aims to estimate emissions on n_point sources, from a series of measures of particulate matter PM₁₀ concentration, taken as a set of m_monitoring stations of air quality (Peña, Hernández, & Toro, 2010(b)). The solution structure or individual, will possess a fitness function which incorporates a lagrangian gaussian puff model based on the principles of a backward gaussian puff tracking model (Martín Ll., Gonzalez, & Palomino, 2002), (Aceña, Martín, DePascual, Sanfelix, Gomar, & Monfort, 2007), (Israelsson, Do, & Adams, 2006), while the dynamic between emission sources and m_monitoring stations of air quality will be determined by a self-organizing map (SOM) (Isazi V. & Galván L., 2004). As a result of EDAs evolutionary process, the proposed model would yield a series of scattering patterns of learning and development, which will determine the spatial and temporal behavior for particulate matter concentration in an area of study. Keywords: Self Organization Map (SOM), AEPuff Model, lagrangian gaussian puff model (LGPM), backward gaussian puff tracking (BGPT), particulate matter PM₁₀, Evolutionary Algorithms, Estimation Distribution Algorithms EDAs.

Hoor, Peter M.

Transport Times in the ExUTLS: Does Static Stability Matter?

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The extratropical upper troposphere lower stratosphere (ExUTLS) has been identified as a transition region between

the troposphere and stratosphere. It contains the tropopause inversion layer (TIL) which constitutes a region of enhanced static stability above the thermal tropopause. Furthermore, the region can be characterized by a transition of transit times from short and rapid processes close to the troposphere to longer time scales deeper in the lowermost stratosphere. Three major transport processes determine the structure of the ExUTLS region: The large scale Brewer-Dobson circulation transports stratospheric aged air down to the tropopause region. Second, the ExUTLS is strongly affected by rapid and frequent exchange across the extratropical tropopause. A third important pathway constitutes quasi horizontal transport from the tropics to high latitudes in the tropical controlled layer above the tropical tropopause. These processes have different time scales and seasonalities affecting the composition in different ways. We will present results of comprehensive backward trajectory calculations of 90 days and 270 days, respectively, to investigate the temporal structure of the lower stratosphere. We will demonstrate, that the key quantity, which determines the tracer abundance in the stratosphere is time since tropopause crossing. Our results indicate, that the time since tropopause crossing is also intimately related to the static stability in the extratropical UTLS which therefore directly affects the depths of the layer influenced by extratropical cross tropopause exchange.

Jaiswal, Nitin K.

Particulate Source Apportionment in Central India using Hybrid Single-Particle Lagrangian Integrated Trajectory Model

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The carbon containing coarse and fine air borne particulate matters (PM) are primary products of combustion, industrial or natural processes, and secondary products of gas to particle conversion in the atmosphere. They have been extensively studied due to their adverse health and climate effects. There are three types of carbonaceous species: carbonate carbon (CC), organic carbon (OC) and elemental carbon (EC) and, they are partially or completely mixed with the PM. The PM can exert a negative radiative forcing of the same magnitude as the greenhouse gases (GHG) and cause cooling effect. The extent and variability of carbonaceous particulate air pollution in the city of Raipur, Central India, surrounded by heavy industries has been investigated. The potential source contribution function (PSCF) analysis model was used for the apportionment of the distant sources. Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPPLIT4) model developed by National Oceanic Atmospheric Administration (NOAA) was used to estimate the air parcel path starting from Raipur city. Hourly five-day backward trajectories starting at 500 m height were estimated using global

reanalysis data. The total PM₁₀, PM_{2.5}, EC₁₀, EC_{2.5}, OC₁₀ and OC_{2.5} concentrations measured at the city of Raipur were used in the PSCF analysis.

Joeckel, Patrick

Lagrangian Transport in a Chemistry Climate Model: Methods and Applications

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The global ECHAM5/MESy Atmospheric Chemistry (EMAC) Model is equipped with the Lagrangian transport submodel ATTLA and further corresponding Lagrangian process and diagnostic submodels. The long-term objective of this development is a full representation of atmospheric chemistry related processes in the Lagrangian frame of reference on the global scale. Furthermore, the system serves as a test-bed for new approaches, like the development of a Lagrangian dynamical climate model core. Despite the common advantages of the Lagrangian approach (mainly mass conservation and non-diffusivity), it provides valuable additional information about spatio-temporal interrelations of atmospheric constituents, which are hardly available from Eulerian models. We present our methods for inter-particle diffusion, large scale and convective transport. The results shown focus on the mass exchange between the stratosphere and the troposphere, on lower stratospheric age of air spectra, and on an inter-comparison of Eulerian and Lagrangian convection and their influence on vertical tracer gradients.

Jones, Andrew R.

Towards a probabilistic approach for hazard area assessment in dispersion modelling

Jones, Andrew R.¹

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Ensemble Prediction Systems (EPS) have become an important tool in operational weather forecasting for assessing uncertainty in meteorological predictions. They provide guidance on the general level of confidence in the forecast evolution (e.g. in support of deterministic products), help to identify possible alternative outcomes (including high-impact weather events) and facilitate a probabilistic approach to weather prediction. Direct model output from EPS forecasting systems is now being deployed in downstream models and decision-making tools (e.g. for flood forecasting applications, or in energy-demand models). The Met Office has been exploring the use of real-time EPS forecasts in its Lagrangian atmospheric dispersion model, NAME. The capability has been developed to run NAME using the ECMWF EPS for applications at medium-range timescales (3 to 10 days ahead) and the Met Office MOGREPS forecasting system at shorter range (0 to 2 days). These capabilities are presently being evaluated, with a specific focus on emergency-response type dispersion

applications. Using ensemble NWP forecasts to drive dispersion models allows us to quantify the effects on the dispersion prediction of uncertainty in the meteorological forecast. Here the EPS approach provides a natural conceptual framework for representing meteorological uncertainties in the dispersion predictions. However, it is also important to acknowledge that other sources of uncertainty exist in dispersion problems, which are often comparable in magnitude (or larger than) the meteorological uncertainty. For instance, large uncertainties often surround the specification of source terms. There are also limitations and errors inherent in the dispersion models themselves. Identifying and understanding these other uncertainties, and then implementing a suitable framework for their representation (e.g. using Monte-Carlo methods), are challenging tasks. Of equal challenge to modelling the uncertainties is how to then communicate this uncertainty information to decision makers so that guidance can be used effectively to make well-informed and sound judgements. Ensemble NWP systems produce estimates of the forecast distributions of the modelled variables. Various ensemble products can be presented from these distributions, including estimates for the probability of a prescribed event, percentiles of the distribution, as well as other statistical measures such as ensemble mean and spread. Similar products can be generated from dispersion ensembles, however one needs to ask whether it is sensible to talk about quantitative “probabilities” when some of the underlying uncertainties in the modelling may be poorly understood. Perhaps it is better here to instead use terms such as “level of confidence” or “level of model agreement”? This presentation will review our experiences with using EPS forecasts in the NAME dispersion model. The focus will be on modelling an “area at risk”, which is often a key concern for decision makers in emergency situations. For example this might be defined as an area where the concentration or dosage is predicted to exceed some intervention level or protection threshold. Various uncertainty products will be presented and their strengths and weaknesses at providing effective guidance to decision makers for hazard area assessment will be discussed.

Joos, Hanna

A Lagrangian climatology of warm conveyor belts over the past 20 years

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During their intensification and in the mature stage, extratropical cyclones are typically associated with three coherent major airstreams – the dry intrusion, the cold conveyor belt and the warm conveyor belt, respectively. These airstreams are important for the dynamics of the cyclone evolution, and potentially also for the rapid long-range transport of atmospheric constituents. Climatologically, they are key for the meridional and vertical transport of

water vapor and heat, linking the atmospheric boundary layer and the tropopause region. Warm Conveyor Belts (WCBs) are strongly ascending cloud producing flows. During their rapid ascent from the boundary layer to the upper troposphere in about 1-2 days, they transport a large quantity of sensible and latent heat poleward and upwards, which leads to cloud formation, the release of latent heat and a significant modification of tropospheric potential vorticity. In this study a detailed Lagrangian climatology of WCBs is compiled for the 20-year time period of the ERA-Interim reanalysis data set (1989-2009). WCBs are identified from comprehensive trajectory calculations that select air parcels in the vicinity of cyclones with a minimum ascent of 600 hPa in 48 hours. The global geographical distribution of WCB starting regions and their tracks will be presented for different seasons. A central point of the study will be the analysis of the typical evolution of key parameters along the WCB flows, including water vapor, cloud water content (liquid and ice), potential temperature, and potential vorticity. It will be investigated whether the typical PV evolution along WCBs is sensitive to the geographical region, the season, the initial moisture content, and the cloud evolution along the ascent.

Joos, Hanna

Microphysical processes and the associated latent heating along trajectories in an extra-tropical cyclone

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Extra-tropical cyclones are of great importance for the weather and climate in the mid-latitudes. They are typically associated with three coherent air streams characterized by different properties, namely a cold conveyor belt, a dry intrusion and a warm conveyor belt (WCB). The WCB describes a strongly ascending airstream that originates in the atmospheric boundary layer in the warm sector of the cyclone. While ascending, clouds form and latent heat is released which intensifies the upward motion. The air parcels typically ascend about 600 hPa in the time range of 48 h and finally reach the upper troposphere. The WCB therefore connects the different tropospheric sub-layers and is responsible for most of the precipitation occurring in cyclones. In this study we use a Lagrangian analysis in order to investigate the evolution of microphysical properties and the associated latent heat release in WCB trajectories. The limited area model COSMO is used in order to simulate a cyclone that occurred in January 2009 in the eastern North Atlantic. In this simulation, the latent heat release due to different microphysical processes occurring during the formation of clouds in the WCB is calculated with a high output frequency of 15 minutes. Based on the model output, WCB trajectories are calculated with the LAGRangian ANalysis TOol LAGRANTO and the different hydrometeor species as well as the latent heating/cooling rates are tracked along the trajectories. As the WCB ascends from the surface to the upper troposphere, different hydrometeor species

form along the ascending airparcels. Close to the surface, cloud droplets and rain are seen by the airparcels whereas in the upper troposphere snow and ice start to form. The complex phase transitions that are associated with the formation of the different hydrometeor species lead to a variety of heating and cooling rates, respectively. Thus, the air parcels experience e.g. cooled due to the evaporation of rain or melting of snow and heating due to the condensation of water vapor or depositional growth of snow. With the Lagrangian analysis presented here, it is possible to investigate the evolution of microphysical properties and the latent heating/cooling along the main airstreams in extratropical cyclones and their influence on the cyclones' development.

Karstens, Ute

Regional-scale atmospheric inversions of greenhouse gas fluxes in Europe

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Estimating regional-scale surface fluxes of long-lived trace gases from atmospheric concentration measurements requires the use of a high-resolution inversion system capable of accounting for small-scale variability in fluxes and transport within the domain as well as large-scale influences penetrating the domain through the lateral boundaries. A two-step nesting scheme was developed, which is based on a decomposition of the atmospheric concentration signal into a near-field component originating from a region and time period of interest and a far-field component supplying information from data outside the region. This nesting scheme allows the use of completely independent models for the representation of the global and the regional transport. Here we use the coupled system TM3-STILT, a combination of the regional Stochastic Time-Inverted Lagrangian Transport model STILT and the global 3-dimensional transport model TM3. The regional TM3-STILT inversion system is applied to estimate surface fluxes of CH₄ and N₂O in Europe. The inversions combine information from hourly atmospheric concentration measurements at 8-10 European continental sites and flask/hourly measurements at a large number of global sites together with a-priori fluxes obtained from emission inventories. European fluxes are estimated for 2006-2007 with a nominal spatial resolution of up to 0.25° x 0.25°. Sensitivity studies will be presented to illustrate the capabilities of the regional inversion system and the potential improvement due to the high resolution in transport and fluxes.

Kaufmann, Pirmin

Sensitivity of wet deposition to the interpolation of meteorological fields in the offline coupled dispersion model Flexpart

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Meteorological fields defined at discrete points in space and time are normally used to drive Lagrangian dispersion simulations. The way the interpolation of these fields is done however may have a appreciable impact on the outcome of the simulation. For example, wet deposition of aerosols often critically determines the deposition pattern of radionuclides released in simulated nuclear accidents. Since wet deposition depends strongly and non-linearly on precipitation intensity, it is no surprise that the way precipitation is interpolated from the meteorological model to the offline coupled dispersion model can have a significant impact on the wet deposition field. In this study, the dispersion model FLEXPART is adapted to use meteorological data from the mesoscale model COSMO. FLEXPART is a freely available Lagrangian particle dispersion model used at many institutes throughout Europe. COSMO is the numerical weather prediction and climate simulation model of a consortium of European weather services and universities. While the original FLEXPART, optimized for the use with global models, assumes that only part of a grid cell is affected by precipitation, and linearly interpolates in time, these choices might not be optimal for convection resolving models. The sensitivity of the wet deposition to the interpolation of the precipitation field is investigated and different options are discussed in terms of physical rationality. The role of cloud coverage and its derivation either from the relative humidity or from the respective parameterized field of the meteorological model is investigated. The impact of different choices for the interpolation on the simulated deposition fields is demonstrated with case studies carried out for nuclear emergency response scenarios.

Keller, Christoph

Inverse Modeling of European Halogenated Greenhouse Gas Emissions Based on Atmospheric Observations and the Lagrangian Particle Dispersion Model FLEXPART

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Anthropogenic halogenated hydrocarbons are potent greenhouse gases which are used in a wide range of applications such as air conditioning, refrigeration, or foam blowing. Official emission inventories of these compounds, if existing, are compiled from statistical data on production and consumption (bottom-up) and often have inherent uncertainties. Therefore, an independent validation of these numbers by top down approaches is essential. In this study, we use continuous air measurements at three long-term monitoring sites (Jungfraujoch, Switzerland; Mace Head, Ireland; Monte Cimone, Italy) in combination with calculations from the atmospheric dispersion model FLEXPART to assess European halocarbon emissions for the year 2009. To better constrain Eastern European emissions, we performed a half-year measurement campaign at a rural site in Hungary (K-Puszta) and integrated these data into our analysis. This resulted in a significant extension of the inversion domain towards Eastern Europe, and it also helped to better constrain Central European emissions as these become tracked from an additional angle. We find that chlorofluorocarbons (CFC) emissions still account for approx. 30% of total halocarbon emissions in Europe, pointing towards large amounts of banked CFCs. The highest current emission rates are obtained for the hydrofluorocarbons (HFCs), which have been employed as substitutes for the CFCs and whose emissions are regulated in the Kyoto Protocol. Our derived values for Western Europe compare well against the official inventories, while the inferred Eastern European emissions are significantly above the numbers reported to the United Nations Framework Convention on Climate Change (UNFCCC).

Kim, Jooil

Measurements of Greenhouse Gases at Gosan (Jeju Island, Korea) for Regional Analysis of Emissions in East Asia

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3. Met Office, Exeter, United Kingdom
4. Analytical and Testing Center, Yanbian University, Yanji, China
5. Scripps Institution of Oceanography, UC San Diego, La Jolla, CA, USA

The large emissions of greenhouse gases (GHGs) from East Asia remain poorly understood. Emission estimation based on actual atmospheric measurements (so-called top-down methods) can help greatly reduce the uncertainties associated with this region. High-frequency *in situ* measurements of GHGs at Gosan (Jeju Island, Korea) capture both clean baseline and pollution plumes arriving from the surrounding region, helping to diagnose the emissions occurring in East Asia. Currently measured species include CO₂, CO, CH₄, O₃, SO₂, and NO_x, as well as a variety of halogenated compounds (CFCs, HCFCs, HFCs, PFCs, SF₆) under the Advanced Global Atmospheric Gases Experiment (AGAGE). Lagrangian modeling of air masses arriving at Gosan have been used to identify periods of dominant influence from China, Korea, Japan, and Taiwan, helping to understand emission characteristics from each of these countries. Baseline conditions at Gosan are well-matched with periods of transport of clean air from northern Asia (in winter) or the Pacific Ocean from the south (in summer), also reflected in the Lagrangian back-trajectory modeling results. Simple source-receptor relationships have been used to estimate the potential source regions and relative emission strengths of CO₂ pollution events observed at Gosan. Recently, more advanced inverse methods utilizing Lagrangian modeling have been applied to the Gosan measurements of halogenated compounds, revealing large emissions of these compounds and suggesting that much of the emissions of these compounds in East Asia are underestimated even in the latest emission inventories. Current inversion modeling efforts have been focused on quantifying the emissions of Perfluorinated Carbons (PFCs) in East Asia, occurring from the aluminum production and semiconductor/LCD manufacturing industries dominant in this region, using both an *a priori* based inversion scheme using FLEXPART and a more numerical approach based on NAME.

Kirwan, A. D.

The 3-D Lagrangian Description of Ocean Eddies

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The application of geometric or dynamical systems methods such as attracting and repelling material manifolds along with their surrogates finite time/space Lyapunov exponents provide geophysical fluid dynamicists with powerful tools for studying advective transport processes. To date most oceanic applications have been confined to the near surface region, and with the tacit assumption of negligible vertical velocity. Typical products are material curves that delineate intricate transport pathways in mesoscale eddy fields. However, these curves are really just intersections of two-dimensional time dependent material manifolds with horizontal levels. Little is known about the vertical extent of these special surfaces. Here we review preliminary results from a systematic study in an eddy-rich region from the Gulf of Mexico. We show that these structures can be traced to several hundred meters depth, and that they delineate nearly columnar 3D “leaky” eddies. Examples of exchange between a large Gulf of Mexico ring and its environment via a turnstile lobe mechanism, and on the vertical material structure of the Loop Current boundary are presented.

<http://www.ceoe.udel.edu/people/profile.aspx?adk>

Kirwan, A. D.

A Lagrangian Perspective of the Deepwater Horizon Spill

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We report on an extensive *a posteriori* analysis of forecasts of the movement of the surface plume from the Macondo oil well disaster in the Gulf of Mexico on 10 April 2010. The analysis uses near surface velocities from a data assimilating regional circulation model. Two metrics are introduced to measure forecast success: percent of the predicted spill area contained in the observations of the spill, and percent of the observed spill contained in the forecast. Passive advection of the spill produces 30 – 75% of the forecast in the observations, while the second metric scores around 30%. The role of wind was found to be negligible except possibly near the shoreline. Several measures of Lagrangian coherent structures are assessed for their capability to describe plume advection and are found to have some skill in capturing the deformation patterns seen in the observations. Finally we comment on steps needed to improve the Lagrangian predictability of ocean circulation models.

<http://www.ceoe.udel.edu/people/profile.aspx?adk>

Konopka, Paul

Diabatic versus kinematic trajectory modeling of stratospheric transport

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We present Lagrangian backward trajectory calculations to analyze the impact of the choice of the vertical transport representation on modeling transport in the tropical stratosphere. Therefore, a diabatic and a kinematic vertical transport representation are used, with the vertical velocity either deduced from the diabatic heat budget (diabatic) or diagnosed from the imbalance of the horizontal winds via the continuity equation (kinematic). The wind data for the calculation is either from ECMWF ERA-Interim reanalysis or from the General Circulation Model (GCM) WACCM. We find that throughout the tropics diabatic transport is homogeneously directed upwards whereas kinematic transport shows large-scale regions of downward motion and a much higher small-scale noise in the vertical velocity field. Both effects yield a much higher vertical dispersion of kinematic than diabatic trajectories. For ERA-Interim reanalysis data, with kinematic velocities influenced by the assimilation, the differences between diabatic and kinematic transport are rather large. But the differences are significant even for GCM meteorological data without assimilation effects. Finally, the diabatic and kinematic backward trajectories are used to reconstruct water vapour and ozone mixing ratios in the tropical lower stratosphere, with the reconstruction method based on either freeze-drying due to the Lagrangian temperature history for water vapor, or on photochemical production and transport for ozone. We show that the uncertainty due to the choice of diabatic or kinematic vertical transport affects the transport of trace gases, in particular for species with distinct spatial gradients like ozone. In general, the larger trajectory dispersion causes significantly higher tropical ozone mixing ratios for kinematic compared to diabatic transport.

Konopka, Paul

Transport in the Chemical Lagrangian Model of the Stratosphere (CLaMS)

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Lagrangian transport has gained increasing popularity in the last few decades not only within the atmospheric community. The chance to avoid, or at least to minimize, the numerical diffusion ever-present in the Eulerian numerical schemes is to date the strongest motivation for the Lagrangian formulation of transport. Supported by new developments in the numerical methods for irregular grids (e.g., $\tilde{\Delta}$ Delaunay triangulation) which are decisive for every

Lagrangian-based numerics, we show in this paper the basic concepts and numerical requirements constraining both the Eulerian and Lagrangian approach. Using the Chemical Lagrangian Model of the Stratosphere (CLaMS), which was developed over the last 14 years, we give some examples how Lagrangian-based transport works in the real atmosphere and why such or similar concepts may replace the traditional, Eulerian-based approach. CLaMS is a Chemistry Transport Model that was successfully applied both to quantify the ozone loss in the stratosphere and to study the stratosphere-troposphere exchange. In particular, we show how the full Lagrangian view offers a possibility to exploit the numerical diffusion for parameterization of the physical mixing rather than to find ways how to avoid this effect. Although mixing at the molecular scale is accomplished by small-scale processes such as turbulence and molecular diffusion, large-scale dynamics can be viewed as the driving factor for these sub-grid processes. CLaMS takes advantage of this physical connection between large-scale and small-scale processes and couples mixing between the Lagrangian air parcels with the large-scale deformations in the flow driven by the vertical shear and horizontal strain on spatial scales of the order 100 km. In particular, the horizontal and vertical scales of mixing are realized in terms of appropriate horizontal and vertical resolutions of the numerical grid. Focusing on the Lagrangian (irregular) grid we derive consistent horizontal and vertical scales from the assumption that their ratio, the so-called aspect ratio, is controlled by the static stability and from the assumption that the entropy of the system is uniformly distributed over all air parcels.

Kretschmer, Roberto

Uncertainty of simulated vertical mixing of CO₂ within the Lagrangian transport model framework WRF-STILT-VPRM

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Atmospheric measurements of CO₂ concentrations over continents, in the proximity of sources/sinks at the earth's surface, contain valuable information to constrain associated processes in the carbon cycle. In the so-called top-down approach, such observations are combined with inverse transport models to estimate surface-atmosphere exchange fluxes. The spatially and temporally highly variable signals put strong demands on these transport models, as they have to represent mesoscale transport phenomena adequately in order to derive accurate flux estimates. Previous studies indicate potentially large model errors associated to the vertical mixing of CO₂ within the Planetary Boundary Layer (PBL). In our study we investigate the impact of uncertain PBL mixing on simulated CO₂ transport. We simulated a time period during the growing season with the Stochastic Time Inverted Lagrangian Transport model (STILT) driven by winds calculated using the Weather Research and Forecasting model (WRF)

employing different PBL parameterizations. To isolate the effect of differences in the transport we prescribed the same CO₂ fluxes for all our simulations that were calculated with the satellite data driven Vegetation Photosynthesis and Respiration diagnostic model (VPRM). Results strongly indicate that observations of the mixing height, i.e. the height up to which tracers in the lower atmosphere are mixed on hourly time scales, can provide crucial information to reduce transport errors related to vertical mixing. Using a pseudo-data experiment, we assess the potential of propagating the transport related uncertainty using the STILT model.

Kunz, Anne

Atmospheric transport pathways near the jet streams and their impact on the UT/LS trace gas distribution

Kunz, Anne¹; Pan, Laura L.²; Konopka, Paul¹; Kinnison, Douglas E.²; Tilmes, Simone²

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Recent analyses of potential vorticity (PV) and trace gas fields on middle world isentropes have shown an overall consistence of the dynamical and the chemical discontinuity at the extratropical tropopause. In particular, in the vicinity of the tropopause enhanced isentropic PV and trace gas gradients are associated with the subtropical and polar jet streams. These analyses motivate the concept of a PV-gradient based tropopause. Atmospheric transport pathways near the jet streams, influencing the transport and mixing of trace gases across the PV-gradient based tropopause, are the focus of this study. These transport pathways are analyzed on the one hand using in-situ observed O₃ and CO during the START08 campaign in April 2008. A mixing layer containing both tropospheric and stratospheric air mass properties is observed near the subtropical jet stream. This mixing layer is located around the PV-gradient based and the thermal tropopauses. Preferred pathways across the tropopause on isentropes above, across and below the subtropical jet stream reveal prescribed branches in the mixing layer based on O₃ and CO correlations. Using the global chemistry model WACCM, we found a pronounced seasonal cycle of the mixing layer around the PV-gradient based tropopause and the location of the transport pathways relative to the jet streams, when the tropopause becomes a stronger transport barrier from summer to winter. Backward trajectories are calculated using the Lagrangian model CLaMS, which are initialized at the PV-gradient based tropopause on middle world isentropes throughout the year. The seasonal evolution of the transport pathways are characterized to better understand their contributions to stratosphere-troposphere exchange.

Legras, Bernard

Lagrangian Transport in the Tropical Tropopause Layer and Convective Sources (*INVITED*)

Legras, Bernard¹; Tzella, Alexandra¹

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We review the application of Lagrangian transport to the Tropical Tropopause Layer and the dehydration of air as it enters the stratosphere. We emphasize the role of exchanges with the extratropical lower stratosphere and of accounting properly of vertical transport by heating rates in the TTL. In particular, we show that the heating provided by high-altitude clouds is important to bridge the gap between the level of detrainment from clouds and that of ascending motion in clear air. It is also found that the convective sources contributing to inject air in the TTL are unevenly distributed among tropical clouds and that the transit times across the TTL exhibit a fat tail due to parcels wandering near the level of zero radiative heating.

www.lmd.ens.fr/legras

Liberato, Margarida L.

The Role of Extratropical Cyclones on the Transport of Atmospheric Water Vapour

Liberato, Margarida L.^{1,2}; Nieto, Raquel³; Trigo, Isabel F.^{1,4}; Durán-Quesada, Ana M.³; Ramos, Alexandre M.³; Trigo, Ricardo M.¹; Gimeno, Luis³

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Recent works point out that moisture contribution for the heavy and moderated precipitation does not come from total column water vapour (TCWV, also known as precipitable water) or from evaporation, but has to come from moisture convergence by the storm-scale circulation (Trenberth et al. 2003). It is thus crucial to know the main sources of the moisture that became precipitation in a given region, distinguishing which moisture is associated with large distance transport and with the low levels convergence. Storms, in particular extratropical cyclones, reach out to gather water vapour over regions that are 10–25 times as large as the precipitation area. This paper aims at characterizing the role of extratropical cyclones on the transport of atmospheric water vapour as well as at identifying the climatological main moisture sources over the Euro-Atlantic region through the application of a Lagrangian methodology to a cyclone climatology. In order to perform a detailed analysis of moisture sources and its associated transport near Iberia, we make use of the method developed by Stohl and James (2004, 2005) which relies on the Lagrangian particle dispersion model FLEXPART (Stohl et al. 2005). Using this model, the atmosphere is initially

homogeneously divided into a large number of virtual particles which have a constant mass and are then advected by the model through three-dimensional wind fields as well as superimposed stochastic turbulent and convective motions. Particle positions and specific humidity information are recorded every specified time step; once the trajectories are computed, moisture variations along the trajectories are calculated by means of specific humidity changes in time. We make also use of a SLP based cyclone detecting and tracking methodology to obtain climatologies of midlatitude cyclones over the North Atlantic European sector (85°W-50°E; 20°N-70°N). This is based on the cyclone detecting and tracking algorithm first developed for the Mediterranean region (Trigo et al. 1999) and later extended to a larger Euro-Atlantic region (Trigo 2006). The scheme is applied to 6-hourly geopotential data at 1000-hPa from ERA40 reanalyses datasets provided by the ECMWF (European Centre for Medium-Range Weather Forecasts), spanning the extended winter seasons from October 1979 to March 2000 and using the highest spatial resolution available. We will focus, in particular, on a number of selected case studies corresponding to heavy precipitation events occurred in western Iberia. Finally the variability of identified moisture sources is analyzed and how these are affected by the large-scale atmospheric circulation patterns will be discussed. Stohl A, P James (2004) *J. Hydrometeorol.* 5: 656–678 Stohl A, P James (2005) *J. Hydrometeorol.* 6: 961–984 Stohl A, C Forster, A Frank, P Seibert, G Wotawa (2005) *Atmos. Chem. Phys.* 5: 2461–2474 Trenberth KE, Dai A, Rasmussen RM, Parsons DB (2003) *Bull Am Meteorol Soc.* 84: 1205–1217 Trigo IF, TD Davies, GR Bigg (1999) *J. Climate* 12: 1685–1696 Trigo IF (2006) *Clim. Dyn.* 26: 127–143 This work was partially supported by the FCT (Portugal) through project PEst-OE/CTE/LA0019/2011-IDL and by CRUP (Portugal) through project No. E-163/10.

Lin, John C.

A backward-time Lagrangian air quality modelling system

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2. Air Resources Laboratory, NOAA, Silver Spring, MD, USA
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We introduce a time-reversed Lagrangian air quality modelling system. The system incorporates a Lagrangian particle dispersion model (STILT/HYSPLIT) running backward in time from receptor sites, along with an atmospheric chemical solver (CB4), as well as gridded emissions of chemical species. Boundary conditions derive from a global chemical transport model (GEOS-Chem). The time-reversed formulation facilitates examination of the meteorological and chemical influences on air quality at the receptor site and enables quick evaluation of the impact of different emission scenarios. We illustrate the application of

the modelling system to simulating air quality in Ontario, Canada.

Malo, Alain

Application of the Canadian Atmospheric Lagrangian Particle Transport and Dispersion Model MLDP0 to the May-June 2010 Forest Fires Episode in Central Québec: A Case Study for Quantitative Validations

Malo, Alain¹; Servranckx, René¹; Bensimon, Dov¹; Bourgouin, Pierre¹; D'Amours, Réal^{1,2}; Ek, Nils¹; Zaganescu, Calin¹; Trudel, Serge¹; Gauthier, Jean-Philippe¹; Mercier, Gilles¹; Hogue, Richard¹

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2. University of Alberta, Edmonton, AB, Canada

Environment Canada's (EC) Environmental Emergency Response (EER) Section at the Canadian Meteorological Centre (CMC) is responsible for the provision of atmospheric transport and dispersion (ATD) modelling products to various clients and partners. The EER Section operates on a 24/7 basis for national and international mandates supporting lead response organizations for environmental emergencies. The team is able to respond quickly and efficiently through well-defined operational procedures for requesting services and via an advanced graphical tool box for providing modelling support. Over the years, the EER Section has developed several ATD models covering a wide range of spatial and temporal scales: long-range (global), medium-range (meso-scale), short-range (local) and urban scale. All atmospheric models can be launched through the operational tool box and are tightly coupled with CMC's numerical weather prediction (NWP) model, the Global Environmental Multiscale (GEM) model. Among the ATD models operated by the EER team is the long-range Lagrangian particle dispersion model MLDP0 (French acronym for *Modèle Lagrangien de Dispersion de Particules d'ordre zéro*) running operationally at CMC. The model is used extensively by Montréal RSMC (Regional Specialized Meteorological Centre) and Montréal VAAC (Volcanic Ash Advisory Centre) staff as well as by the EER modellers and duty officers on a regular basis as part of their operational mandates. We briefly describe the MLDP0 model and present an example of its application to the May-June 2010 forest fires episode in central Québec, Canada. Numerical forward simulations were performed for multiple sources using MLDP0 driven by analyzed meteorological data from the GEM NWP Regional model at 15 km horizontal grid mesh. The modelling results were compared as time series against PM_{2.5} measurements for several stations. Observations were made available by two air quality monitoring networks: the RSQA (French acronym for *Réseau de surveillance de la qualité de l'air de la Ville de Montréal*) and the MDDEP (French acronym for *Ministère du Développement durable, Environnement et Parcs du Québec*). The large plumes generated by the forest fires together with the abundance of high-quality surface pollutant measurements from many air quality monitoring stations covering a wide geographical

area, provided an exceptional opportunity to 1) illustrate the usefulness of the model in assessing the motion and dispersion of forest fire plumes over medium and large scales, 2) quantitatively validate the model against observations, and 3) estimate total PM_{2.5} quantities released by the forest fires.

Miller, John B.

Forward and Inverse Modeling of CO₂ and δ¹³C at North American Tall Towers using the FLEXPART Lagrangian Model

Miller, John B.¹

1. R/GMD1, NOAA/ESRL, Boulder, CO, USA

Historically, ¹³C has been used to partition CO₂ fluxes into land and ocean components, because terrestrial CO₂ fluxes (both photosynthesis and respiration) impose distinctive patterns upon the atmospheric ¹³C ratio. However, as measurement density increases, ¹³C has the potential for an important new role in carbon cycle research: to explore mechanisms driving terrestrial biosphere exchange variability. In particular, in regions dominated by terrestrial fluxes, the atmospheric δ¹³C signal can be dominated by stomatal conductance, which is an extremely important parameter in understanding fluxes of water, energy and carbon (and thus, both climate and weather research). Here we will present results of forward and inverse modeling of year-long time series of CO₂ and δ¹³C at two North American tall towers: LEF (Wisconsin) and AMT (Maine). First, we use the FLEXPART Lagrangian Particle Dispersion Model (LPDM) to predict CO₂ mole fractions ¹³C ratios at the tower sites using a full specification of the CO₂ and ¹³C budgets added to an advected background contribution. The inverse modeling component of our study then involves minimizing the differences between observed and predicted mixing and isotopic ratios by adjust both terrestrial fluxes and their isotopic ratios at a variety of temporal and spatial scales. Preliminary results from forward modeling at LEF show very good agreement between observed and predicted CO₂. In fact, using CarbonTracker a posteriori terrestrial fluxes as input to our regional Lagrangian system yields better agreement with observations than CarbonTracker itself. This suggests that “near-field” transport in FLEXPART is more accurate than in the Eulerian model TM5 (that drives CarbonTracker) and/or the advection of North Pacific boundary air is more accurate in our regional framework than that global Eulerian CarbonTracker framework. Using this initial result as motivation, we will present results of tests designed to disentangle these two hypotheses.

Miller, Scot M.

Constraints on nitrous oxide and methane sources over North America and their underlying drivers

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4. Earth Systems Research Laboratory, Global Monitoring Division, NOAA, Boulder, CO, USA

We present top-down constraints on nitrous oxide and methane sources over the United States and Canada, respectively, and provide insight into the underlying processes driving emissions. We analyze data from aircraft measurements and tall towers between 2004 and 2008 using a high resolution Lagrangian particle dispersion model known as STILT applied to both geostatistical and Bayesian inversion frameworks. Our results indicate strongly seasonal N₂O emissions that peak in June and closely mirror the spatial distribution of fertilizer application. We find a large methane source over the Hudson Bay Lowland in Canada, which exhibits a seasonality different from most existing inventories. Our analysis suggests a need to improve the treatment of soil temperature in many biogeochemical models of wetland methane sources.

Miltenberger, Annette K.

Lagrangian Perspective on Warm-Rain Orographic Precipitation in Idealized Three-Dimensional Simulations

Miltenberger, Annette K.¹; Joos, Hanna¹; Seifert, Axel²; Wernli, Heini¹

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2. Deutscher Wetterdienst, Offenbach, Germany

Due to its meteorological and societal importance, orographic precipitation is a classical theme of meteorology, but the involved interactions of nonlinear dynamics and microphysics still pose challenges. In recent studies the basic mechanisms have been investigated based on the characteristic timescales of orographic precipitation formation. However, these timescales are poorly constrained by observations and theoretical considerations. Investigations of numerical simulations from a Lagrangian perspective can provide deeper insights in the underlying physical mechanisms that determine these timescales. In this study, the usefulness and the potential of the Lagrangian perspective is demonstrated in a trajectory-based investigation of warm-rain stable orographic precipitation at an isolated mountain simulated with the non-hydrostatic COSMO model. The study focused on two essential timescales of warm-rain clouds: The advective timescale,

describing the time air parcels spent inside the cloud, and the microphysical timescale, describing the time required to form rain. It is shown that different flow configurations lead to different cloud structures that are associated with significant variability of the two timescales. This is related to the gravity wave structure and interactions of microphysical and dynamical processes. Such variations were suspected, but could not be quantified in earlier studies. Based on the trajectory data timescales characterizing entire clouds have been calculated. Their dependence on the vertical Froude number shows significant and robust deviations from previous theoretical formulations: The advective timescale decreases less strongly than expected with increasing Froude number, which is related to the influence of the gravity wave on lee side cloud extent. The rain formation induces in addition a dependence on the microphysical conditions not acknowledged in previous work. The microphysical timescale increases stronger than expected with decreasing Froude number for low flow velocities. The different behavior can be assigned to a larger reduction of vertical velocities than predicted by geometrical considerations due to the onset of lateral flow deflection. This idealized study of an archetypal atmospheric flow configuration illustrates the potential of the Lagrangian approach in leading to novel insight into the associated dynamical and microphysical processes and their interaction.

Minejima, Chika

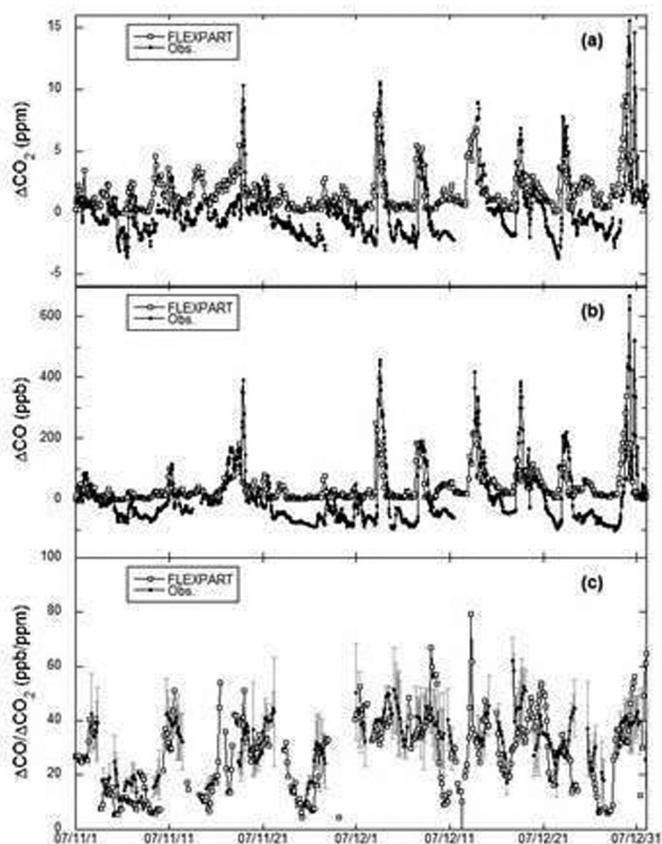
Comparisons of observed and simulated CO₂, CO, CH₄, and O₂ at Hateruma Island, JAPAN, by using FLEXPART and global coupled Eulerian-Lagrangian transport model

Minejima, Chika¹; Tohjima, Yasunori²; Yamagishi, Hiroaki²; Koyama, Yuji²; Maksyutov, Shamil²; Tanimoto, Hiroshi²; Machida, Toshinobu²; Mukai, Hitoshi²

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2. NIES, Tsukuba, Japan

In-situ observations of atmospheric CO₂, O₂, CO and CH₄ concentrations at Hateruma Island (HAT, 24°N, 124°E) often show synoptic scale variations reflecting air masses transported from East and South East Asian source/sink regions. The time series of these variations show noticeable similarity during the period from late autumn to early spring. We had calculated the correlation slopes of the two components for the highly variable events with the duration of a few days, and found that the slopes show significant differences depending on the air mass origins categorized by backward trajectories (e.g. Tohjima et al., ACP, 2010; Minejima et al., ACPD, 2011). In order to deepen our understanding of the synoptic scale variations, we simulate the atmospheric concentrations by using FLEXPART (Stohl et al., Atmos. Environ., 1998) and a global coupled Eulerian-Lagrangian transport model (Koyama et al., GMD, 2011) with a range of surface fluxes of CO₂, O₂, CO, and CH₄. The simulated variations show good agreement with the observations, especially phases. Fig. 1 shows the time series

of the simulated and observed CO₂ and CO for Nov.-Dec. 2007. The amplitude of the simulated variations is generally underestimated in comparison with the observations. We calculate the CO/CO₂ correlation slopes for the observed and simulated time series for the moving 24-hour time window by using RMA regression (Fig. 1c). The observed CO/CO₂ slopes show significant variations, reflecting the facts that (1) there is heterogeneity in the regional distribution of the CO/CO₂ emission ratios (higher CO/CO₂ ratio in China than in Japan/Korea) and (2) the contribution of the regional emissions to the observation at HAT varies with time. The simulated CO/CO₂ slopes agree well with the observation, suggesting the model used in this study fairly well reproduce the relative contributions of the regional emissions. We will compare the correlation slopes of the other pairs of the above species between observation and simulation more in details at the conference.



The time series of the detrended and deseasonalized (a) CO₂ and (b) CO concentrations at HAT, and (c) the CO/CO₂ correlation slopes.

Mohanakumar, Kesavapillai

Exchange of Water Vapor from Troposphere to Stratosphere During Asian Summer Monsoon Season

Mohanakumar, Kesavapillai¹

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Asian summer monsoon has distinct effects on the upper troposphere and lower stratosphere. It exhibits strong

interannual variability which is controlled by several local and remote processes, and intra-seasonal variability, that is manifested by active and break phases. This study explores the inter-annual and intra-seasonal variability of water vapor in the troposphere and its transport to the lower stratosphere during the summer season over the Asian monsoon region. About 75% of the water vapor transport in the upper troposphere of the Asian summer monsoon region is contributed by the monsoon circulation from June to September. The inter-annual variations of upper tropospheric humidity (UTH) correspond well with those of rainfall showing a clear reduction in 2002 and 2009, years of weak monsoons in the last decade. Average residence time of water vapor in the atmosphere is around few days. Drying of the upper troposphere over the Arabian Sea in 2002, which was a result of an unusually long monsoon break from end of June to middle of July associated with dry air from Arabia capping the convection above the boundary layer. In contrast, 2009 exhibited a much larger scale upper tropospheric drying all over the monsoon region almost throughout the season. The impact of moisture on large scale intra-seasonal variability can be understood as a feedback process that involves moist convection, UTH and the large scale flow. Knowledge of the distribution and variability of upper tropospheric humidity can, therefore, give insight into the mechanisms of monsoon variability.

Mortarini, Luca

Lagrangian Particle Model Approach to Two-Phase Releases. The MicroSpray New Module

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3. SIMULARIA S.r.l., Torino, Italy

The Lagrangian Stochastic approach is among the most suitable for fast emergency response models dealing with hazardous releases in real case scenarios. In this framework the simulation of the dispersion of aerosols clouds forming from accidental ruptures in pressurized vessels containing toxic industrial chemicals is of particular importance. For this purpose a new version of the Lagrangian dispersion model MicroSpray was developed. The two-phase dynamics is modelled taking into account the density effects and the thermodynamic transformations assuming homogeneous thermodynamic equilibrium at each time step. A system of differential equations is solved for each stochastic particle to simultaneously evaluate the dynamic and the thermodynamic of the plume. After the initial flashing the evolution of the liquid and vapour mass fraction of the contaminant, of the water vapour and dry air is evaluated. The negative buoyancy effect is modelled coupling a modified set of Glendening equations with the energy conservation and state equations. The model details and some tests of its capabilities will be shown: a sensitivity analysis of the new module for the simulation of flashing

jets, field experiments from the Modelers Data Archive and a chlorine railway accident (Macdona) will be presented.

Nehrkorn, Thomas

Lagrangian Modeling of Greenhouse Gas Observations in Urban Environments

Nehrkorn, Thomas¹; McKain, Kathryn²; Wofsy, Steven²; Henderson, John¹; Leidner, Mark¹; Ellis, Marikate¹; Eluszkiewicz, Janusz¹; Levi, Patricia²

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2. Harvard University, Cambridge, MA, USA

Quantifying current and modeling future emissions of CO₂ from urban areas are important components of climate change research and monitoring. Accurate estimates of CO₂ emissions require a combination of approaches: using inventories of known anthropogenic and natural sources (“bottom-up” approach); analyzing flux measurements at selected sites; and combining concentration measurements with transport and dispersion computations to constrain prior estimates of sources (“top-down” approach). We are reporting on applications of the Stochastic Time-Inverted Lagrangian Transport (STILT) model driven by meteorological fields from the Weather Research and Forecasting (WRF) model for the modeling of greenhouse gas concentrations in urban environments. Results will be presented to illustrate the role of different components of the modeling framework that affect its ability to reproduce observed concentrations at measurement sites located within the urban boundary layer. Key components include the emission inventory, aspects of the WRF model such as spatial resolution, parameterization packages for the planetary boundary layer and urban canopy, and STILT options that control the effect of boundary layer processes on the computation of Lagrangian particle trajectories. Results will be presented from simulations in two very different urban environments: Salt Lake City, Utah, a moderately sized city situated in a mountainous region characterized by relatively sparse vegetation, and Boston, Massachusetts, a large metropolitan area in a coastal location with extensive forested areas upwind. This research builds upon and extends other efforts to develop and apply the WRF-STILT model to regional top-down GHG flux estimates, with other examples to be given in the companion paper by Eluszkiewicz et al. We gratefully acknowledge funding by U.S. Government Agencies.

Novakovskaia, Elena

Sensitivity of STILT footprints to assimilation of weather observations within the transport model

Novakovskaia, Elena¹

1. Earth Networks, Inc., Germantown, MD, USA

Over the next five years, Earth Networks will deploy and operate a network of Greenhouse Gas (GHG) measuring instruments installed at tall towers. Continuous observations of atmospheric carbon dioxide and methane mixing ratios provided by this network will be used for

inverse modeling to estimate natural and anthropogenic sources and sinks of GHGs at regional and local scales. For these inversions, the atmospheric trajectories and surface footprints are computed using the STILT (Stochastic Time-Inverted Lagrangian Transport) model coupled to the WRF (Weather Research and Forecasting), which provides fine spatial and temporal resolution atmospheric transport fields at local scale. Errors in simulated meteorological transport fields are commonly considered as the dominant source of errors and uncertainties in estimated carbon fluxes. To improve accuracy of the atmospheric transport model, meteorological observations from the dense surface network operated by Earth Networks are assimilated into the WRF. These weather observations are available for assimilation in real time. Also, data from a few wind profilers are used, which in combination with surface observations improve accuracy of the weather model as shown in the WRF model verification. Trajectories and footprints based on transport fields with and without data assimilation are analyzed to evaluate the sensitivity to corrections in transport fields. It is shown that differences in upstream influences will impact results of inversions suggesting that assimilation of surface weather observations is a necessary step to improve accuracy in estimated sources and sinks of GHGs.

Novitsky, Mikhail A.

The Lagrange Model of an Accidental Release Transport in an Atmosphere with Limitation of Cross-section Component of a Wind Speed Fluctuation

Novitsky, Mikhail A.¹

1. TYPHOON, Obninsk, Russian Federation

Absence of pseudo diffusion is the well-known advantage of Lagrange models of pollutants transport. Disadvantage of such models is necessity of use of the big number of particles at calculations if it is necessary to calculate pollutants concentration. It is convenient to use Lagrange model in calculations of an accidental release transport in which are taken into account limitation of cross-section component of a wind speed fluctuation. Such model has been created for the forecast of pollution of Norilsk city in Russia. In prognostic system for the Norilsk city environment [1] the Euler model for calculation of pollution transport has been replaced on the Lagrange model. Values of maximum fluctuation of wind direction during limited periods of time are obtained on the basis of measurements at a meteorological tower in Obninsk [2]. These periods correspond to the periods of transport pollutant puff at distances about 10 km from a source. Wind direction and speed were measured with a discretion of 1 s at altitudes up to 300 m with their subsequent averaging over 10 s. The average values of maximum fluctuation of wind direction decrease with an increase in wind speed, with a grows of altitude of measurements, and with a rise of atmospheric stability. The data obtained used for estimation of the distribution function in the Lagrange Monte-Carlo model of pollution transport. References 1. V.A. Savchenko

e.a. The system for forecasting the contamination levels from SO₂ industrial emissions for the Noril'sk city environment. 3rd International Symposium on Environmental Hydraulics with a Special Theme in Urban Fluid Dynamics. Tempe, Arizona, USA, December 5-8, 2001. 2. M.A. Novitsky e.a. Maximum fluctuations of wind direction over limited time intervals at altitudes up to 300 m from observations at a meteorological tower. *Meteorologiya i gidrologiya* (in Russian), 2007, No. 3, pp.33-42.

<http://typhoon-tower.obninsk.org>

Orza, José A.

The Influence of NAO on the Interannual Variability of Tropospheric Transport Pathways in Southern Europe

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We present an exploratory study of the interannual variations in tropospheric transport pathways in southern Europe and its relationship to the North Atlantic Oscillation (NAO). A significant fraction of the year-to-year variability in the Northern Hemisphere is associated with this pattern of pressure anomalies, which is typically characterized by the NAO index derived from the difference in surface pressure levels between Iceland and the Azores/Lisbon (Hurrell, 1995). This work relies on the identification of flow types by cluster analysis of back-trajectories and the association between variations in the NAO index and the frequency of occurrence of the identified flow types. Four sites in southern Europe have been selected: two locations in the western Mediterranean basin (Malaga, S Spain; Elche, SE Spain), one in central Mediterranean (Lecce, SE Italy) and one in the Atlantic façade (Izki, N Spain). In addition, one site in central Europe (Cabauw, The Netherlands) has been considered for comparison purposes. For each site, 3D back-trajectories were computed with the HYSPLIT model v4.8 (Draxler & Hess, 2004) and the NCEP/NCAR Global Reanalysis meteorological data, to compute 96-hour back-trajectories, for 4 times each day at 3 heights between surface and 700 hPa. Flow patterns were identified by cluster analysis of backward trajectories spanning the 8-year period 2000 – 2007. The clustering methodology is the one already followed in Dueñas et al (2011). Between 6 and 8 flow types, depending on the location, were identified by the cluster analysis. Clear seasonal patterns are observed. An association is found between the changes in the identified synoptic flow patterns and the NAO for the winter months (DJFM), from the monthly means of the NAO index and the monthly frequencies of the flow patterns. There are common features shared amongst the study sites in southern Europe: Both the frequency of zonal flows and their wind speed are reduced in the positive phase of the NAO, while polar maritime air masses with relatively high winds become more common. Besides, easterlies/north-easterlies are enhanced in the

Mediterranean locations, as the Azores high blocks the entrance of westerlies and polar disturbances. The reduction in westerly/northwesterly flows arriving at 3000 masl is higher than at lower heights during the NAO positive phase in the Mediterranean, due to the strong decoupling of the flows below and above the ABL. Similarly, the increase in the frequency of Mediterranean flows is higher near the surface. While changes in pathways over time overall reflect known synoptic situations, the use of air parcel trajectories provides more detailed information of the pathways; this is useful to identify variations in potential source regions of airborne pollutants or variations in the wind energy resource at a location. M. Cabello acknowledges financial support from the TTORCH Project. Draxler RR, Hess GD (2004). Description of the HYSPLIT_4 modeling system. NOAA Technical Memorandum ERL ARL-224. Dueñas C, Orza JAG, Cabello M, Fernandez MC, Cañete S, Perez M, Gordo E (2011). Atmos. Res. 101, 205–214. Hurrell JW (1995). Science 269, 676-679.

Pan, Laura

Chemical Transport and Mixing near the Tropopause: Integrated Studies using Chemical Tracer Measurements and Lagrangian Models *(INVITED)*

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The chemical composition in the tropopause region, particularly the gradient of several radiatively sensitive species, is of importance to chemistry-climate interactions. Two-way exchange and mixing of stratospheric and tropospheric air is a major process that controls the tracer gradient in this region. Lagrangian models are important tools for these process studies. We present two case studies to address the issues of how well the small scale mixing can be parameterized using large scale flows and the possible representation of convective transport using trajectory models and wind fields from global analyses. The first case study demonstrates the use of the Chemical Lagrangian Model of the Stratosphere (CLaMS), integrated with the aircraft in situ observations during the Stratosphere Troposphere Analyses of Regional Transport 2008 (START08) experiment, to understand the formation of the Extratropical Transition Layer (ExTL). In this case, tracer-tracer correlation is shown to be an essential tool for integrating models and observations. The second case study focuses on the issues of air mass origins and the role of convective transport in the region of the Asian Summer Monsoon, using the TRAJ3D model, together with ozone and water vapor measurements. In this case, we discuss the impact of using different meteorological analyses. Together, these case studies show that Lagrangian models have their

unique strength in integrating tracer observations with dynamical processes and in quantifying transport and mixing.

Pillai, Dhanyalekshmi

Towards regional carbon budgeting using WRF/STILT-VPRM: A comparison of Lagrangian and Eulerian models for atmospheric CO₂ transport

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Atmospheric CO₂ concentrations provided by two high-resolution modeling systems – the Eulerian and Lagrangian transport models – are compared as a step towards Bayesian inverse modeling technique to retrieve regional carbon budgets. The modeling system consists of a Eulerian-based transport model – the Weather Research Forecasting (WRF) – and a Lagrangian-based transport model – the Stochastic Time-Inverted Lagrangian Transport (STILT). Both transport models are coupled to a satellite-based biospheric model – the Vegetation Photosynthesis and Respiration Model (VPRM) to generate atmospheric CO₂ concentration in the atmosphere. A quantitative comparison between the two different approaches, while using identical surface-atmosphere fluxes and meteorological fields, is a prerequisite for applying STILT as an adjoint of WRF for inverse modeling. The consistency of the simulations is assessed with special attention paid to the details of horizontal as well as vertical transport and mixing of CO₂ concentrations in the atmosphere. A case study using airborne measurements during which both models showed large deviations is analyzed in detail as an extreme case. Using aircraft observations and pulse release simulations, we identified differences in the representation of details in the interaction between turbulent mixing and advection through wind shear as the main cause of discrepancies between WRF and STILT transport. Based on observations and inter-model comparisons of atmospheric CO₂ concentrations, it is shown that a further refinement of the parameterization of turbulent velocity variance and Lagrangian time-scale in STILT is needed to achieve a better match between the Eulerian and the Lagrangian transport at such a high spatial resolution. Nevertheless, the inter-model differences in simulated CO₂ time series for a tall tower observatory at Ochsenkopf in Germany (50° 1'48"N N, 11° 48'30" E) are about a factor of two smaller than the model-data mismatch and about a factor of three smaller than the mismatch between the current global model simulations and the data, which justifies using STILT as an adjoint transport model of WRF.

Pisso, Ignacio J.

Emission location dependent ozone depletion potentials for very short-lived halogenated species

Pisso, Ignacio J.¹; Yang, Xin²; Law, Kathy³; Haynes, Peter²

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2. University of Cambridge, Cambridge, United Kingdom
3. LATMOS, Paris, France

We present trajectory-based estimates of Ozone Depletion Potentials (ODPs) for very short-lived halogenated source gases as a function of surface emission location. The ODPs are determined by the fraction of source gas and its degradation products which reach the stratosphere, depending primarily on tropospheric transport and chemistry, and the effect of the resulting reactive halogen in the stratosphere, which is determined by stratospheric transport and chemistry, in particular by stratospheric residence time. Reflecting the different timescales and physico-chemical processes in the troposphere and stratosphere, the estimates are based on calculation of separate ensembles of trajectories for the troposphere and stratosphere. A methodology is described by which information from the two ensembles can be combined to give the ODPs. The ODP estimates for a species with a fixed 20 d lifetime, representing a compound like n-propyl bromide, are presented as an example. The estimated ODPs show strong geographical and seasonal variation, particularly within the tropics. The values of the ODPs are sensitive to the inclusion of a convective parametrization in the trajectory calculations, but the relative spatial and seasonal variation is not. The results imply that ODPs are largest for emissions from South and South-East Asia during Northern Hemisphere summer and from the Western Pacific during Northern Hemisphere winter. Large ODPs are also estimated for emissions throughout the tropics with non-negligible values also extending into northern mid-latitudes, particularly in the summer. These first estimates, whilst made under some simplifying assumptions, show larger ODPs for certain emission regions, particularly South Asia in NH Summer, than have typically been reported by previous studies for emissions distributed over land in within broad latitudinal bands. We discuss recent developments aiming a more detailed treatment of tropospheric chemistry based on off line coupling with a chemical transport model.

Pisso, Ignacio J.

Constraints on CO₂ flux emissions based on reconstructions of in-situ measurements from Lagrangian stochastic inversion

Pisso, Ignacio J.¹; Patra, Prabir¹; Takigawa, Masayuki¹; Nakazawa, Takakiyo²; Sawa, Yousuke³; Machida, Toshinobu⁴; Matsueda, Hidekazu³

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3. MRI, Tsukuba, Japan
4. NIES, Tsukuba, Japan

In order to use high resolution in-situ measurements to constrain regional emissions of carbon dioxide (CO₂) we use a Lagrangian methodology based on diffusive backward trajectory tracer reconstructions. We use aircraft, ground and tower sites for CO₂ data, collected during the CONTRAIL campaign, from the MRI/JMA Tsukuba tall tower, nearby the CO₂ emission hot spot of the Tokyo Bay Area and from the World Data Centre for Greenhouse Gases (WDCGG). Estimated fluxes for the Tokyo Bay Area for the analyzed period in 2006/2009 range between $4.8 \cdot 10^{-7}$ to $3.45 \cdot 10^{-6}$ kgCO₂/m²s with significant time variations. Sensitivity to simplified boundary layer representations and turbulent mixing representations was studied as well as uncertainties associated with advective transport based on ECMWF reanalysis / WRF mesoscale wind fields, with measurement error and with aggregation error. We investigate the necessity of regularization based on bayesian and nonlinear positivity constraints respect to inversions with no a priori fluxes and the application to independent assessment of publicly available inventory data. Observation targeting in the context of GHG inversions for error reduction is discussed.

Ramos, Alexandre M.

A new circulation type classification based upon Lagrangian air trajectories

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2. Institute for Atmospheric and Climate Science, ETH, Zurich, Switzerland

Techniques for the classification of circulation types are usually based on a small number of meteorological fields like SLP or geopotential height. In particular, they most often do not take into account the 3d-structure of the flow and they focus on a single day, i.e., they neglect the temporal evolution of the flow. In this work we present a new classification method for the northwestern Iberian Peninsula, based on the analysis of backward trajectories arriving in this target area. The aim is to produce a classification of the representative 3d-flow for each day. The backward trajectories were calculated with the Lagrangian particle dispersion model FLEXPART. Data from a global simulation were used, which contains more than 1 million

particle positions and their meteorological properties recorded every six hours over a five-year period (1/12/1999 to 11/30/2004). The classification method consists of several distinct steps: a) First, a horizontal cluster analysis is performed, based upon the longitude and latitude values of the trajectories. A second clustering then considers the height of the trajectories, their distance to the target area, and their values of specific humidity and latitude in order to further reduce the number of clusters per day. b) Specific flow characteristics are computed for every cluster, including the curvature of the flow, and characteristics of the moisture evolution. Each cluster is then characterized by a subset of physical key variables. c) Finally, each classifying variable was separated into several distinct classes (e.g., cyclonic or anticyclonic flow), hence enabling us to characterize each trajectory cluster with an “index” for each classifying variable. As results, we obtain a daily catalog containing information about the air masses that arrive at the target area (trajectory types). In order to analyze if the new classification method is able to identify the intra-seasonal variability in the region, the frequency of the clusters for each season is examined. Moreover a comparison of this new trajectory classification to the “classical” Lamb weather type classification will be made. Additionally, the relationship between circulation types and precipitation is analysed in order to investigate if the new method is able to distinguish the precipitation variability in the studied region. To conclude, several case studies will be presented in order to illustrate and validate the new classification method. The case studies embrace different synoptic patterns that typically affect the northwestern Iberian Peninsula such as: blocking events, heat-waves, cold waves, cut off lows, extra-tropical cyclones, and cold fronts.

Reisin, Tamir G.

Numerical Simulations of Cloud Rise and Cloud Top Determination Following an Explosion Using a Lagrangian Model

Reisin, Tamir G.¹; Pistinner, Shlomi¹

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One of the most important factors in risk assessment following an explosion is the determination of the source term that serves as input for the atmospheric dispersion. In addition to the amount and kind of material released, this source term also includes the spatial distribution of the pollutants. In the simplest Gaussian models (such as Hotspot) the initial pollutants distribution is determined via a simple formula (Church’s formula) that relates the final cloud top height to the amount of explosive material and assumes a certain vertical distribution of the pollutants mass. More sophisticated models provide additional information on the spatial and temporal evolution of the pollutants cloud. A few field experiments were conducted in order to validate those models. Operation Roller-Coaster was a joint US-UK effort in which a series of high explosives tests were conducted at the Toponah Test Range. The explosive

charge weights were 48-726 Kg. These tests were conducted under a variety of meteorological conditions (ranging from stable to unstable conditions). The measurable quantity in these tests was the cloud top height as a function of time. These measurements were carried using either a photographic or a theodolite tracking system. The aim of the present work is to define quantitatively the rather fuzzy term of “cloud top”. To that goal several Roller-Coaster explosions were simulated using a meteorological model (RAMS) in conjunction with a Lagrangian particle model (HYPACT). The RAMS model was initialized with a hot bubble parameterizing the explosion at the beginning of the buoyant stage, a high resolution grid was used. The dynamic fields calculated by RAMS were used to drive the Lagrangian model (HYPACT). The source term to HYPACT are Lagrangian passive particles released at the explosion location. The spatial and temporal distribution of the Lagrangian particles are analyzed for several tests conducted under stable atmospheric conditions. The cloud top derived from these particle distributions agrees well with the field measurements.

Scheiben, Dominik

Lagrangian trajectory analysis of middle atmospheric ozone and water vapor during the sudden stratospheric warming of January 2010

Scheiben, Dominik¹; Straub, Corinne¹; Hocke, Klemens¹; Kämpfer, Niklaus¹

1. University of Bern, Bern, Switzerland

The major sudden stratospheric warming (SSW) of January 2010 was an extreme event when the northern hemisphere polar vortex shifted from the polar region towards Europe. The warming was accompanied by a zonal wind reversal in the stratosphere and mesosphere from westerlies to easterlies. This change in middle atmospheric dynamics had a major influence on locally observed trace gas species such as ozone and water vapor. The latter is often used as a passive tracer for middle atmospheric dynamics because water vapor can be regarded as relatively inert for time periods of a few days. In this study we present middle atmospheric water vapor and ozone measurements observed by ground-based microwave radiometry during the mentioned SSW. The observations were taken from three different locations across Europe, namely from Bern, Onsala and Sodankylä. At each location, an influence of the SSW on the vertical ozone and water vapor profiles was observed and is explained by a trajectory analysis. The trajectories were calculated by LAGRANTO based on the wind fields of the ECMWF operational analysis. Fundamental differences in the vertical trace gas profiles were observed when the general middle atmospheric regime changes between inside-vortex and outside-vortex conditions. Stratospheric ozone depletion was observed at all three locations, but strongest over Bern. The ozone depletion is explained by advection of ozone-poor air from inside the stratospheric polar vortex. Exceptionally high ozone concentrations were observed over Bern shortly after the SSW and are explained by advection of

subtropical, ozone-rich air due to the change in stratospheric dynamics. The observed water vapor measurements correlate well with ECMWF Potential Vorticity (PV) which in turn is an indicator of the polar vortex. In the polar mesosphere, the water vapor mixing ratio increased by about 50 percent during the SSW, which is explained by transport of humid midlatitudinal air. In the midlatitudes, the observed water vapor anomalies were strongest in the stratosphere, where water vapor increased by about 25 percent during the warming. This is explained by advection of relatively humid air from the stratospheric polar-vortex.

Schnadt Poberaj, Christina

Understanding Recent Methane Growth Rate Variability Using a Global Lagrangian Transport Model – First Results

Schnadt Poberaj, Christina¹; Henne, Stephan¹; Brunner, Dominik¹

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Methane is the second most important well-mixed greenhouse gas in terms of radiative forcing after carbon dioxide. We intend to improve the understanding of recent methane growth rate variability focussing on parts of the period of near zero growth rates between 2004 and 2006 and on the latest increase since 2007. In particular, we aim to better understand the roles of the main source categories and quantify their contribution to the global CH₄ budget, investigate the effect of chemistry and transport on CH₄ variability, and study the impact of meteorological variability on tropical and boreal wetland emissions. We apply the particle dispersion model FLEXPART extended with a simplified first order decay chemistry in a global domain filling mode. Surface emissions are taken up from different sources when particles pass through the planetary boundary layer and methane is removed by its primary loss reaction with the hydroxyl radical, by surface deposition and a stratospheric sink. Each particle carries a set of 44 different tracers each representing a different CH₄ source category in 4 different age classes. Within the study, a series of global forward simulations of the period 2004-2008 was carried out. They consist of a reference simulation forced by varying meteorology and emissions describing atmospheric methane as realistically as possible and two sensitivity simulations, in which individual processes, i.e. emissions and/or OH are kept constant. First results for the reference simulation show very good agreement with ground-based CH₄ measurements of the Global Atmosphere Watch (GAW) program for most stations. The high quality of the simulations allows a credible attribution of methane variability to individual source categories and to distinguish between the influences of emissions and long-range transport. In this presentation, we will specifically highlight the influence of boreal and tropical wetland emissions on recent CH₄ growth rate variability in comparison to variations in OH and transport.

Shershakov, Viatcheslav

The model STADIUM as an instrument for estimating radioactivity dispersion in case of a nuclear accident

Shershakov, Viatcheslav¹; Borodin, Ruslan¹

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The paper presents the features of implementation of the model STADIUM used in the Regional Specialized Meteorological Center (RSMC) for addressing the issues related to atmospheric dispersion of radioactivity as part of information support to implementation of the international convention on early warning of nuclear accidents. The STADIUM (STochastic Atmospheric Diffusion Model) is used for modeling atmospheric transport and dispersion of pollutants (radioactive or chemical). The STADIUM is based on the Lagrangian approach, with the turbulent dispersion simulated by the random walk technique (Monte-Carlo method), which allows applying modern parameterizations for turbulent dispersion and deposition processes. Both wet and dry deposition is computed using dry deposition removal rates and in-cloud and below-cloud removal rates by wet deposition. The model accounts for the key features associated with the atmospheric boundary layer instability and non-uniformity and spatial heterogeneity of the underlying surface. The STADIUM provides a set of spatial and temporal fields of air concentration and deposition (dry and wet) of pollutants. Results are presented on application of the model to reconstruct the dispersion of short-lived radionuclides in the early stage of an accident, given no systematic measurements. Techniques and examples are provided for using the model in other emergencies involving environmental contamination (the nuclear test in North Korea on 9 October 2006, the fall of the Proton-M rocket on 6 September 2007 and others). Computational data using the model STADIUM are compared with data of monitoring the radioactive contamination of the ambient air in the Russian Far East (Vladivostok, Yuzhno-Sakhalinsk, Yuzhno-Kurilsk, Petropavlovsk-Kamchatsky) following the accident at the Fukushima-1 nuclear power plant. The monitoring was based on using collection of samples with air filtering systems and horizontal collectors.

Shin, Seoleun

Hamiltonian Particle-Mesh (HPM) methods for numerical modeling of atmospheric flows

Shin, Seoleun¹; Reich, Sebastian¹

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The Hamiltonian Particle-Mesh (HPM) method is an interesting alternative to Eulerian formulation of dynamical cores, especially regarding to climate simulations since it ensures the exact transport and conservation of mass. This method is combined with a symplectic time-stepping that preserves phase volume, which provide also an excellent conservation of energy and circulation for long-term

simulations. We have developed schemes based on the HPM method for the shallow equations on the sphere and a nonhydrostatic vertical slice model. Series of benchmark tests show that the HPM is capable of representing atmospheric flows and preserves energy to a high degree of accuracy in the long-term simulations. Recently we have developed a hydrostatic HPM method and implemented it in a vertical slice model. The performance of the model is tested for the simulation of idealized linear orographic flow in both dry and moist environments. The HPM method is able to capture the fundamental features of the orographic wave and represent the influence of moisture on wave characteristics consistently with theoretical analysis. In future we investigate a redistribution of particles when diabatic processes lead to a convective instability.

Škerlak, Bojan

A global ERA-interim Climatology of Stratosphere-Troposphere Exchange and Identification of Key Physical Processes

Škerlak, Bojan¹; Sprenger, Michael¹; Pfahl, Stephan¹; Sodemann, Harald¹; Wernli, Heini¹

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Stratosphere-troposphere exchange (STE) has an important impact on atmospheric chemistry and potentially also on the climate system. It is thus important to quantify exchange fluxes of air, water vapour, ozone and other chemical constituents on different spatial and temporal scales. Several climatologies of STE have been compiled in the last decades leading to a fairly good quantification and understanding of the geographical distribution of exchange events on the global scale. The physical processes leading to the exchange, however, have mainly been investigated for particular case studies on a regional scale. Therefore the relative importance of the various processes that can modify the air parcels' potential vorticity and lead to STE (e.g., condensational heating, turbulence, and radiation) is still unknown on the global scale. In this study we use the ERA-Interim reanalysis data set (1989-2010) from the European Centre for Medium-Range Weather Forecasts (ECMWF) in order to produce a new trajectory-based climatology of STE. Compared to earlier reanalyses, the increased spatial resolution and the 4D-Var data assimilation scheme constitute two major advances leading to an increased overall quality of the STE climatology. The Lagrangian methodology to identify STE events is based on backward and forward trajectory calculations initialised from a dense grid in the upper troposphere and lower stratosphere and selects air parcels that (i) cross the 2-pvu dynamical tropopause, and (ii) stay longer than a specified threshold residence time of a few days on both sides of the tropopause before and after the STE event, respectively. The investigation will focus on the geographical distribution of upward and downward cross-tropopause fluxes of mass, water vapour, and ozone, on the seasonal cycle of STE, and in particular on those STE events that lead to a rapid transport of stratospheric air masses down to the

atmospheric boundary layer and vice versa. In addition, various meteorological parameters will be tracked along the STE trajectories (e.g., condensational heating rate, cloud water content, indicators of clear air turbulence, etc.) in order to investigate and quantify in a Lagrangian framework the physical processes that are associated with the cross-tropopause exchange processes.

<http://www.iac.ethz.ch/groups/wernli/>

Sodemann, Harald

An intercomparison of Lagrangian methods to diagnose evaporation regions of water vapour and precipitation (*INVITED*)

Sodemann, Harald¹; Wernli, Heini¹

1. Institute for Atmospheric and Climate Science, ETH Zurich, Zürich, Switzerland

The identification of regions where water evaporated from the surface, so called moisture sources, for precipitation or atmospheric water vapour is currently receiving increased attention. Water source information provides insight into the physical coupling between evaporation and precipitation, which is one important ingredient for the occurrence of precipitation extremes. Recently, several diagnostics based on backward trajectories have been proposed in the literature to identify the moisture sources for precipitation, each with own specific assumptions and simplifications. A direct comparison between these different methods has so far not been carried out. Here we present results from an intercomparison of three recently proposed Lagrangian moisture source diagnostics. The ECMWF ERA-Interim data are used as a common dataset. Three-dimensional kinematic backward-trajectories have been calculated using the LAGRANTO model and the Lagrangian Particle Dispersion Model FLEXPART with and without parameterised convection. The different methods are compared for a high-latitude, mid-latitude and a tropical case, to test the influence of convective moisture transport on the diagnostics. Evaporation fields from ERA-Interim as well as the HOAPS data set are used to evaluate the physical plausibility of the identified moisture source regions. The results demonstrate that the methods differ substantially with respect to the distance of the identified moisture sources for specific events, while the spatial location is more consistent. The representation of vertical motion in the trajectory calculation is a decisive factor for all methods. Based on the comparison with simulated and observed evaporation fields, a critical discussion of the underlying assumptions of the various methods is carried out.

Sprenger, Michael

A Lagrangian climatology of orographic blocking

Sprenger, Michael¹; Madonna, Erica¹; Roch, Alexandre¹

1. ETH Zurich, Zurich, Switzerland

Traditionally, orographic blocking is studied with Eulerian methods based on the estimation of the inverse

Froude number or dimensionless mountain height $F=N/HU$, where N is the stratification, H the mountain height and U the upstream velocity. Here we present the Lagrangian perspective of orographic blocking. Meteorological fields are taken from the high-resolution NWP model COSMO, which is operationally run at the Swiss weather service. Winds are taken from a three-year (2000-2002) reanalysis simulation with COSMO. Based on these winds, kinematic forward trajectories are started at a distance of 300 km all around the Alps and at two height levels (750 and 1500 m). The 24-h trajectories are then investigated in their capability to surmount the Alpine barrier. The blocking climatology is separated into three weather classes: westerly flow, northerly flow and southerly flow, the latter being restricted to south Foehn cases. For each class the percentage of trajectories surmounting the Alps and the percentage of air parcels flowing around the Alps is determined. Furthermore, trajectory densities are calculated to show the different air streams which start from selected upstream positions. The blocking frequencies are compared for air streams starting at 70 m and at 1500 m. Finally, the Lagrangian method to identify orographic blocking is compared to the Eulerian one. The advantages of the Lagrangian method are discussed, as well as its limitations.

Sprenger, Michael

The Lagrangian Analysis Tool Lagranto

Sprenger, Michael¹; Wernli, Heini¹

1. ETH Zurich, Zurich, Switzerland

The calculation of kinematic trajectories is numerically not very demanding. Three-dimensional wind fields are required, which are then used to step forward in time the individual air parcels. Here we present the trajectory tool Lagranto for ECMWF meteorological data, which in addition to simple trajectory calculations offers several add-on features. Firstly, Lagranto allows to trace additional meteorological fields along trajectories, hence allowing in-depth analysis of physical processes experienced by the air parcels on their way through the atmosphere. Secondly, trajectories can easily be selected based on position and other meteorological parameters. The selection criteria can flexibly be combined and include: passage through specified regions, ascent and descent by specified amounts, threshold for meteorological parameters,... Thirdly, Lagranto comes with several supportive tools which invite the user to interactively experiment with trajectories. Furthermore, all features of Lagranto are described in an extensive documentation. In this presentation, the capabilities of Lagranto are shown with some case studies. The examples include: identification of warm-conveyor belts, source-receptor studies, and stratosphere-troposphere exchange.

Sprenger, Michael

Lagrangian perspective of stratosphere-troposphere exchange - physical processes associated with the crossing of the extra-tropical tropopause (*INVITED*)

Sprenger, Michael¹

1. ETH Zurich, Zurich, Switzerland

The dynamical tropopause of the extra-tropics, defined as an iso-surface of potential vorticity (PV), acts as a barrier to stratosphere-troposphere exchange (STE). Therefore, any transport across this barrier is necessarily associated with diabatic or frictional processes such as turbulence, radiative forcing or condensational heating. In this presentation, these exchange processes are studied in a Lagrangian perspective. Our aim is to take the STE air parcels' perspective and to move along with them in their journey from the troposphere to the stratosphere (STT), and vice versa for TST. The analysis is based on a Lagrangian one-year climatology of STE for the northern hemisphere. The three-dimensional kinematic trajectories are based upon the ERA-40 wind fields. For each trajectory crossing the tropopause, time, latitude, longitude, pressure and potential temperature are recorded, together with flags separating stratosphere-to-troposphere transport from TST and shallow from deep exchanges, respectively. In a first part, we look at the vertical transport of the air parcels from one sphere into the other. To this aim we look at all deep STT and TST trajectories from one year and consider the evolution of pressure (height), PV and potential temperature along them. It will be shown that it is essential to separate three different steps in an STT event, and correspondingly in an TST event: i) the transport from the lower troposphere to the upper troposphere; ii) the crossing of the tropopause; and iii) the final mixing with the ambient stratospheric air. Additionally, we compare the vertical velocities associated with STT and TST. Secondly, we address the question which dynamical structures are typical for STE. In particular we investigate if the STE air parcels are associated to stratospheric and tropospheric PV streamers, to tropopause folds, to cyclones and anticyclones and atmospheric blocking. To this aim we rely on several objective climatologies of these features. Furthermore, the link to the jet streams is considered. Here, we propose a 'new' look where not only the horizontal position of the STE air parcels relative to the jet streams and to jet streaks is considered, but the whole 3D surrounding of the STE parcels is taken into account. The surrounding is studied several hours before, at and several hours after the crossing of the tropopause. Finally, in third step, we look at the physical processes which allow air parcels to cross the dynamical tropopause. Essentially, this is done by tracing relevant meteorological fields along the STE trajectories. Composites are then built over many STT and TST events, the time origin shifted to the time when the trajectories cross the tropopause. In a first part, we will look at a decomposition of the velocity field into its invariant parts, i.e. into deformation, vorticity and divergence. It will be shown that STE events are particularly

associated with distinct features of deformation and vorticity. The second part will address in greater detail the diabatic processes which go along with STE. We consider in particular clear air turbulence (CAT), condensational heating and cooling and radiative processes.

Stohl, Andreas

Source identification and quantification with Lagrangian model output: strategies with different complexity (*INVITED*)

Stohl, Andreas¹

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Lagrangian models have traditionally been used to locate and to quantify sources of trace substances (e.g., pollutants, dust, radionuclides, volcanic ash, etc.) into the atmosphere. This contribution will review different approaches for that purpose, such as: 1) Case studies using trajectories or particle dispersion model output 2) Statistical analysis of large trajectory data sets (e.g., cluster analysis, potential source contribution analysis, etc.) 3) Statistical analysis of particle dispersion model output 4) Inverse modeling The contribution will review where we stand with these different approaches and will make recommendations for the use of these methods, depending on the problem to be solved.

Talerko, Mykola

Evaluation of Potential and Limitations of the Lagrangian-Eulerian Mesoscale Model for the Reconstruction of Space-Time Features of Contamination Fields in Belarus and Ukraine Caused by the Chernobyl Accident

Talerko, Mykola¹; Garger, Evgeny¹

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The possibilities of the atmospheric transport model LEDI (Lagrangian-Eulerian Diffusion model) for reconstruction of ¹³¹I air and ground contamination field formation in the territory of Ukraine during the initial period of the Chernobyl accident have been investigated. Using of this model which is based on the Lagrangian approach enables to calculate the contamination transport over Ukraine from prolonged source with time-variable parameters. The first step of this work was modelling of ¹³⁷Cs transport and deposition in Ukraine and the comparison of obtained results with available data of soil contamination measurements. It enables to build the release scenario from the accidental unit of the Chernobyl nuclear power plant (taking into account the time variability of intensity and initial vertical distribution of the release) and to assess the large and small scale features of radioactivity deposition field in Ukraine. The ¹³¹I atmospheric transport over the territory of Ukraine was simulated during the first 12 days after the accident (from 26 April to 7 May 1986) using real aerological information and rain measurement

network data. The airborne ¹³¹I concentration and ground deposition fields were calculated for subsequent thyroid dose reconstruction for inhabitants of radioactive contaminated regions. The obtained results are compared with available data of radioiodine daily deposition measurements made at the network of meteorological stations in Ukraine and data of the assessments of ¹³¹I soil contamination obtained from the ¹²⁹I measurements. The same approach was used for the simulation of radioactive contamination of the territory of Belarus. The comparison of modelling results of ¹³⁷Cs and ¹³¹I deposition values for the settlements of Belarus with available cesium and iodine soil measurements data showed a sufficiently good agreement. It was shown that the used model enables to explain mechanisms of formation of a large-scale spots of radioactive contamination (with a linear scale of about dozens kilometres) which were formed due to wet deposition as well as due to the effect of combination of diurnal variations of the transport conditions and the high-elevated prolonged release source (specific for the Chernobyl accident) on dry deposition of nuclides. Though, as the results of radioactive contamination measurements show, there is a considerable heterogeneity of deposition field with scale about 1 km. This variability may be explained by different reasons: impact of local features of underlying surface which wasn't taken into account under simulation, different characteristics of deposited material (e.g., variability of relative input of different physical and chemical forms of radioiodine), a possible input of wet deposition caused by local rains which hasn't been detected with the rainfall gauge network etc. The small-scale deposition field variability is assessed using data of ¹³⁷Cs detailed measurements in the territory of Ukraine. To describe the local variability of radioactive deposition field we used the approach named "the method of local effective depositions".

Thomas, Jennie L.

Photochemical processing during long-range transport using Eulerian and Lagrangian approaches

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3. USA, Germany, France

During summer 2008 pollution plumes of different origin, including boreal fire plumes, anthropogenic plumes, and plumes that consisted of mixture of different source regions were observed by the NASA DC8 (USA), the ATR-42 (France), and the DLR-Falcon (German) aircraft as part of the POLARCAT IPY field campaign. For example, anthropogenic plumes originating from North America and forest fire plumes from Canada were sampled near the

source region and later downwind over Greenland. The origin of plumes will be discussed as well as their initial chemical composition. Two methods are used to examine photochemistry in plumes as they are transported to the Arctic. Lagrangian modelling including chemistry has previously been used to quantify processes (photochemistry, mixing, deposition) during long-range transport (e.g. Real et al., 2007, 2008) based on multiple samplings of the same plume over a period of several days. Lagrangian matches between upwind and downwind aircraft have also been identified in the POLARCAT dataset. Simulations using a photochemical trajectory model, CiTTYCAT, are used to investigate the photochemical evolution of plumes, in a Lagrangian framework, initialising with upwind data and comparing with downwind data. We also use an Eulerian model, WRF-Chem, to simulate photochemistry and transport during the measurement period at high spatial resolution and to determine the impact of fire and anthropogenic emissions on net ozone production over wider spatial scales. We compare the results of the two methodologies and discuss differences between them. For example, photochemical ozone production and destruction rates along trajectories from WRF-chem are compared to results from Lagrangian calculations.

Thomson, David

History of the Lagrangian Stochastic model for turbulent dispersion, Part 2

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This talk will survey the evolution of the Lagrangian stochastic model from the mid 1980's to the present day. (Part 1 covers the period from Taylor (1921) until the early 1980's). The emphasis will be on models where the particle state is defined by the particle's position and velocity, although not exclusively so. The mid 1980's saw a range of ideas appear on how to treat flows where the turbulent velocity scale varies with position or where the velocity distribution is non-Gaussian, and these ideas lead to the principle that if the position-velocity distribution of particles is "well-mixed" it should remain so. Using these ideas led to successes in predicting dispersion in a number of such flows. In particular, a number of authors demonstrated the ability of the resulting models to represent convective boundary layer dispersion. This is difficult using other modelling approaches because of the strongly inhomogeneous and non-Gaussian character of the flow. As a result the convective boundary layer became the application which arguably best demonstrates the advantages of the Lagrangian stochastic technique. In 3-D flows, the well-mixed condition together with a choice of time-scale (or of the constant C_0 in the inertial-subrange Lagrangian velocity structure function) is not sufficient to

determine the formulation of the model uniquely. How to choose the most appropriate model from the range of possible models remains an open problem, although progress has been made in understanding the implications of this uncertainty. In particular it was discovered that the extra freedom available in 3-D makes it possible for the velocity vector to "rotate" systematically. In models where this occurs the rate of dispersion is reduced. The value of C_0 is the second main open problem and a number of studies which made progress in determining the most appropriate value will be discussed. While the models where the particle state is defined by position and velocity form the core of the subject, a number of extensions have been explored. These include models to represent turbulence intermittency, dissipation range properties, stable stratification, deep convection, and inertial or sedimenting particles, as well as models for predicting concentration fluctuations and chemical reactions, such as multi-particle models and models using "interaction with the mean" (or with the conditional mean). At the same time simpler "diffusive" models where the particle state involves only the position have become widely used as cheaper options for representing unresolved motions, e.g. in conjunction with the resolved velocity field from numerical weather prediction models for calculating long range atmospheric dispersion. This wide range of model types will be briefly surveyed.

Tschanz, Brigitte

Analyzing Middle Atmospheric Water Vapor Variations during SSW 2010 using a Lagrangian Trajectory Model

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From January to June 2010 the ground-based microwave radiometer for middle atmospheric water vapor MIAWARA-C took part in the LAPBIAT campaign at the Finnish Meteorological Institute Arctic Research Centre in Sodankylä, Finland. As a result of this campaign an almost uninterrupted time series of water vapor profiles ranging from approximately 32 km to 75 km could be retrieved. During the sudden stratospheric warming in January 2010 MIAWARA-C measured abrupt changes in mesospheric water vapor which are attributed to transport from the middle latitudes. The Lagrangian-based trajectory model LAGRANTO is used to test this hypothesis and to illustrate transport processes during a SSW. The measured water vapor profiles are compared to profiles determined by calculating backward trajectories.

Tsunematsu, Kae

Modeling transport of volcanic particles with a multiparticle cellular automata method

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Explosive volcanic eruptions produce a range of particle sizes that are injected into the atmosphere and are transported by the wind until they eventually sediment on the ground at various distances from the vent. Modeling the transport and sedimentation of volcanic particles is crucial to the assessment of associated hazards, such as disruption to aviation, crop pollution, damage to lifelines and collapse of buildings. Generally transport of volcanic particles in the atmosphere is treated as an advection-diffusion process. In fact, particles are advected by wind and affected by the diffusion due to turbulence in the atmosphere and in the plume. Separately from the volcanological approach, numerical models for advection-diffusion are studied by using the cellular automata (CA) method and the Lattice Boltzmann (LB) method. Especially, a multiparticle CA model, which calculate the trajectory of each particle traveling on a grid system, successfully reproduced transport of snow and sand particles [2]. We have found that multiparticle CA is more efficient than the LB approach in terms of memory and CPU time [3]. Thus, we have applied this multiparticle CA model to simulate the transport of volcanic particles with appropriate boundary conditions. A fundamental issue in particle dispersal models is the characterization of the source term [1]. Our model includes the plume velocity field obtained by extending the 1D plume model [4] to 3D. With this simplified velocity field of the plume, our model successfully reproduce the source term of particle release. Besides, computed ground sedimentation shows a good agreement with field observations from selected Pululagua and Cotopaxi eruptions. Our model can be applied for forecasting both ground sedimentation and concentration of particles in the atmosphere, which is useful for aviation industry, and it is easy to parallelize efficiently on large scale parallel machines. References: [1] Bonadonna, C, Folch, A, Loughlin, S, Puempel, H (2011), "Ash Dispersal Forecast and Civil Aviation Workshop - Consensual Document," <https://vhub.org/resources/503> [2] Dupuis, A, and B Chopard.,(2002), Lattice gas modeling of scour formation under submarine pipelines. *J. Comp. Phys.*, 178:161–174. [3] Tsunematsu, K, J-L, Falcone, C, Bonadonna and B,Chopard, (2008) Applying a Cellular Automata Method for the Study of Transport and Deposition of Volcanic Particles, *LNCS 5191*, 393-400. [4] Woods, A W, (1988), The fluid dynamics and thermodynamics of eruption columns, *Bull. Volcanol.*, 50, 169–193.

Ubl, Sandy

Exploring Atmospheric Transport of PCBs Measured in the Arctic

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Polychlorinated biphenyls (PCBs) are persistent organic pollutants (POPs) which are assumed to have no significant natural sources. PCBs are regulated globally under the Stockholm Convention on POPs. PCBs are toxic, persistent, bioaccumulative and undergo long-range transport in the environment. However, their long range transport potential is not yet fully understood; an important factor is the degree of chlorination, which in turn determines the PCBs volatility and reactivity with OH radicals. Here, we investigate the role of atmospheric transport on the PCB concentrations measured in the Arctic. We combined PCB concentrations measured in air at the monitoring sites Birkenes, Zeppelin and Alert with calculations using FLEXPART, a Lagrangian Particle Dispersion Model. The focus is on the Arctic sites, Alert and Zeppelin, and Birkenes is chosen as a reference station where it is known that measured and modeled PCB concentrations are in good agreement. We use existing backward model runs for the above mentioned stations which range 20 days backward in time and calculated an emission sensitivity (ES) function. In winter, air originating from Europe, Russia and Asia will arrive at Alert and Zeppelin and even PCB sources located in southern Europe can contribute to measured PCB concentrations at these stations. PCB sources located in the northern part of Europe, Asia and Canada will contribute to measured PCB concentrations in summer. However, highest ES values are found over the Arctic ocean and decrease sharply near the continental coasts. Modeled and measured concentrations agree reasonably well at Birkenes. Highest correlation coefficients, r , are found in summer for PCB 28 and PCB 52 with $r=0.77$ and $r=0.8$, respectively. Heavier congeners like PCB 118, 138, 153 and 180 correlate only in summer with correlation coefficients ranging from $r=0.56$ to $r=0.38$. In comparison, appreciable correlation at Zeppelin is only found in autumn and winter for PCB 101 with $r=0.56$ and $r=0.42$. For heavier congeners, r is around $r=0.3$. There is no significant correlation for the lighter congeners. At Alert, correlation is generally low, with a maximum of $r=0.4$ for PCB 101 in spring. PCB 28 is the only congener with significant correlation for all seasons, although $r=0.3$ is very low. The reasonable agreement of modeled and measured PCB concentration at Zeppelin and especially at Birkenes indicates that primary emissions together with atmospheric transport determine the variability of PCB concentration measured at these stations. Additionally it points to the suitability of FLEXPART studying long range transport potential of PCBs. However, regarding the poor agreement at Alert, other processes that are not covered by the model are

dominating. We therefore will compare the FLEXPART model results with results derived from a multimedia model to investigate the relevant processes.

Vermeulen, Alexander T.

Spatial Estimates for Methane and Nitrous Oxide Emissions at the Continental Scale Using a Direct Inversion Technique With Recursive Source Area Aggregation

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The representativity of mixing ratio observations in the boundary layer is an important issue and depends for example on the location of the site and its climatology, sources and sink strength in the immediate surroundings, sampling strategy and the observed component. Depending on the component, area and processes one wishes to study the sampling strategy will therefore vary strongly. For long lived greenhouse gases with high background mixing ratio values and relatively small sources (and sinks, if any) this will work out completely different as for short lived reactive tracers like ozone or terpenes. In all cases the variability through emissions or reactions at time scales of seconds to hours will be observed together with variability due to atmospheric transport and mixing processes on these same time scales. On longer time scales of days to months the mixing ratio signal observed is always a composed signal due to processes on connected larger spatial scales from continental to hemispheric scale. On the annual to climatic time scales global processes will start to dominate the variations. Disentangling the different processes at different timescales requires the use of atmospheric transport models that are sufficiently adequate at the required scales in time and space. A combination of coupled transport models each working at different characteristic scales in time and space seems to be a feasible way to carry out analysis of observed time series at discrete observation sites. In this paper we present a combination of the offline coupled eulerian global transport model TM5, providing the global boundary conditions, and a set of Eulerian and Lagrangian regional transport models. From this combination we derive by direct inversion a spatial explicit emission map for methane and nitrous oxide in western Europe, based on continuous mixing ratio observations of these gases at a set of background and tall tower sites in Europe. The regional models used are the eulerian WRF model and the lagrangian models Flexpart and COMET. The inverted emission maps vary in resolution from 10*10 km close to the observation points to 1000*1000 km at large distances from the observations. Total emissions for methane on a country by country comparison with bottom up estimates compare generally well, although significant differences larger than the uncertainty estimates are found. The differences between

inversion results and bottom-up inventory nitrous oxide, where the largest uncertainty in emissions is expected, are smaller than for methane.

<http://www.ecn.nl>

Vogel, Baerbel

Using peroxyacetyl nitrate (PAN) - tracer correlations in the tropopause region to quantify stratosphere-troposphere exchange in Lagrangian model simulations

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Peroxyacetyl nitrate (PAN; CH₃C(O)OONO₂) is mainly generated in air masses polluted by combustion of fossil fuels and by biomass burning and has a lifetime in the lower troposphere in the order of hours. However in the upper troposphere, PAN has a longer lifetime of the order of several weeks, because the thermal decomposition of PAN is slow. In this region, UV photolysis and reaction with OH can become important loss processes for PAN. Thus PAN can be transported throughout the cold upper troposphere over long distances and is therefore an excellent tracer of polluted air masses transported into the lower stratosphere upper troposphere region (UTLS). It can be used to study (1) mixing processes in the UTLS and (2) different transport pathways into the UTLS. The Lagrangian 3-dimensional chemistry transport model CLaMS uses 3-dimensional deformations of the large-scale winds to parameterize mixing. Several previous studies showed that first, CLaMS has the ability to characterize mixing near the tropopause and second, transport pathways can be identified using tracers of air mass origin. This motivates us to incorporate PAN chemistry in CLaMS using results of an air quality model as lower boundary condition (= PAN sources). Sensitivity studies will be performed analyzing PAN - ozone correlations derived from CLaMS simulations, aircraft campaigns, and satellite measurements to quantify stratosphere-troposphere exchange.

Vogel, Felix R.

Potentials (and problems) of validating atmospheric transport and emission modeling of fossil fuel CO₂ and other tracers at an urban site by in-situ observations

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Lagrangian back-trajectory models are commonly used to relate regional observations to fluxes from specific emitters or regions. To be able to assess their ability to correctly model the atmospheric concentrations of Greenhouse Gases (GHGs) they need to be tested e.g. utilizing tracers with well-known flux fields. For fossil fuel CO₂ (FFCO₂) emission data is available from high spatial resolution inventories. Thus the emergence of proxy-based hourly FFCO₂ observations from urban sites might open the door for an assessment of common model problems, such as mixing within the nocturnal stable layer. This could be evaluated using hourly FFCO₂ observational data from urban sites where the FFCO₂ signal is very pronounced. This might furthermore help to identify other wrongly implemented processes in our framework. This study uses the multi-year 14C-calibrated CO-based FFCO₂ observations, as well as data of numerous other trace gases (222Rn, CH₄, N₂O) in the urban environment of Heidelberg, Germany. The in-situ observations of 2005 and 2006 are compared with simulations from the Stochastic Time-Inverted Lagrangian Transport model (STILT), using 25kmx25km ECMWF meteorology. The anthropogenic emissions of GHGs are taken from a new fine-gridded emission model from IER (2005) that provides sector-specific hourly fluxes at 5' by 5' resolution for Europe. Besides comparing the modeled and the measured time-series during special episodes, a more aggregated way of comparison is proposed by using mean diurnal cycles of different tracers for all seasons. Additional information is gained by also comparing the model results for other tracers (CH₄, N₂O, CO) with the local measurements. Comparing STILT-IER2005 results with 14C-calibrated CO-based FFCO₂ reveals that STILT accumulates too much tracer during the night-time for all modeled GHGs. Using a tracer with homogenous emissions such as 222Rn or others can help to quantify this effect. Furthermore our study showed that the linkage of emission model and transport model has to be revisited as the emissions from stacks are only parameterized using a single emission height. This can lead to a strong overestimate of the influence of specific emission sectors in Lagrangian models. To improve the performance

of the Lagrangian modeling framework, the emissions of GHGs have to be included in a real 4-D manner. Furthermore, the influence from meteorology on emissions could be taken into account, e.g. the temperature dependent emissions from specific sectors (i.e. domestic heating). At last a model calibration-strategy using FFCO₂ and other tracers such as 222Rn should be implemented. It is also suggested to include FFCO₂ in studies that are focusing on other tracers so that FFCO₂ can be used for a quality assessment of the simulation.

Wang, Junming

PM10 contribution from agricultural tilling operations

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It is very important for management purposes to know how different sources contribute to regional particulate matter (PM) and what the contribution percentages are, especially the dust sources from agricultural operations. Regional PM10 (PM of aerodynamic diameter $\leq 10 \mu\text{m}$) contributions from agricultural tilling operations were estimated from Hysplit4 model and airplane PM10 measurements in the Mesilla Valley, Las Cruces, NM. A Dustrack PM10 sampler on an airplane measured regional dust concentration at a height of 200 m with 1-s frequency on April 1, 2008. Hysplit4 model was run at the corresponding time and height and outputted the PM10 concentrations from all assumed agricultural tilling operations. The measured PM10 concentration was then used as the total regional PM10 and was compared with the simulated measurements from all the simultaneous tilling operations from the Las Cruces area. The average PM10 contribution (0.0009 mg/m³, with a standard deviation 0.0008 mg/m³) from tilling operations was about 19% of the average measured PM10 concentration (0.0048 mg/m³, with a standard deviation 0.0017 mg/m³).

Wang, Kuoying

Assessment of industrial air pollution episodes with high resolution Lagrangian model

Wang, Kuoying¹

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Efficient and accurate attribution of sources and emission strengths are key steps to reconstruct processes leading to air pollution episodes, and to conduct air pollution impact assessment. During heavy industrial air pollution episodes, we need to be able to quickly provide scientifically credible results that are simple but clear enough to convince both the industries on the scope of

pollution been emitted and its impacts on the environment; and to the general public on the temporal-spatial scale of air pollution episodes. These are not easy tasks, which requires timely and accurate monitoring of ambient air pollutions and meteorology, and the capability to link monitoring time series data to air pollution sources and strength. In this work we develop a 100-m resolution Lagrangian model to assess air pollution complaints made by the general public against a major oil refinery park located in Northern Taiwan. A total of 101 complaints were made during 2008-2009 from locations within 3-km radius of the oil refinery park. The Lagrangian model was systematically run through each complaint with real observed winds to study the relationship between the pollution sources, transport, and the arrival of the model emissions at the location and the time when the complaints were made. We found that the Lagrangian model is very effective in converting the complex relationship of winds and pollutant dispersions into results that can be understood both by the industry and the public. The Lagrangian model has also been validated against severe air pollution episodes arise from industrial accidents and forest fires. These results demonstrate the value of the Lagrangian model in real-world air pollution episodes assessments. More validations are continuously been searched and tested to further consolidate the accuracy of the Lagrangian model in industrial air pollution episode assessment.

<http://140.115.35.249>

Webley, Peter

Improving the Accuracy of Eruption Source Parameters for Aviation Safety

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Volcanic ash clouds represent a major hazard to aircraft. Since 1953, more than 130 aircraft worldwide have reported encounters with ash and at least 79 have sustained damage. In 1987 the International Civil Aviation Organization (ICAO) responded to the threat of ash clouds by creating the International Airways Volcano Watch, whose purpose was to develop global procedures to disseminate information and warnings of ash cloud hazards to aviation. In the early 1990s ICAO established nine Volcanic Ash Advisory Centers (VAACs) whose purpose is to monitor and forecast the movement of ash clouds in their region and to issue advisories to the aviation community. To forecast ash-cloud movement, VAACs use transport and dispersion models, whose accuracy is limited largely by incomplete knowledge of eruption source parameters such as plume height, start and stop time, mass eruption rate, and the mass fraction of the erupted material consisting of fine ash that feeds the cloud. From 2007-2009, a multidisciplinary group working under

the aegis of ICAO recommended improved methods for estimating source parameters (Mastin et al., 2009a, JGVR 186:10-21), and developed a spreadsheet of likely source parameters for future eruptions at the world's active volcanoes (Mastin et al., 2009b, USGS Open file Report 2009-1133). The accuracy of source parameters (and their variation with time) became more important during the Eyjafjallajökull eruption in April 2010, when changes in flight procedures in Europe required model forecasts to produce maps of ash concentration, delineating zones containing concentrations of 0.2-2, 2-4, and >4 mg m⁻³. In 2010 ICAO established a Volcanic Ash Task Force to evaluate the utility of adopting similar procedures worldwide, and a Volcanic Ash Scientific Advisory Group (VASAG) to advise the Task Force on limitations and possible improvements in model accuracy. Current practices for estimating mass eruption rate during an eruption have an uncertainty of an order of magnitude, meaning that model forecasts cannot resolve small differences in concentration (e.g. between 2 and 4 mg m⁻³ as current European procedures require). Several groups represented in the VASAG are working to improve the real-time accuracy of operational ash-cloud models by (1) developing automated ensemble modeling, (2) integrating satellite, LiDAR, or other observational constraints, or (3) establishing a web-based clearinghouse of source parameter information. Here, we present some of the findings of this work and discuss the usage of the improved ESP data for the volcanic ash Lagrangian models, such as Puff, HYSPLIT, NAME and FLEXPART.

Webley, Peter W.

Pre-eruption warnings of the potential hazards from volcanic ash clouds

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Hazardous volcanic ash clouds can develop quickly and there is a need to forecast their movement to provide advisories to the aviation community. Volcanoes are monitored in real-time to detect for impending activity with warnings based on an aviation 'color code' and advisory/watch/warning alert system. Using this alert system, volcanic ash dispersion and transport (VATD) models can forecast the potential ash cloud even before the event has occurred. This can provide a first stage assessment of the volcanic hazard. Here we present how the Lagrangian based Puff VATD model is used for forecasting ash cloud movement in real-time, pre- and during an eruption. The Puff model tracks each individual ash particle in space and time, advecting and dispersing the particles throughout the atmosphere using multiple numerical weather prediction data as its advection term. Before an eruption, many of the VATD model parameters are unknown and therefore multiple scenarios are required. Most can be obtained from the eruption source parameters work of Mastin et al (2009;

JGVR 186:10-21). The most variable parameter is the initial plume height. The Puff model simulates eruption cloud movement for plumes for over 30 volcanoes worldwide from 4 – 16 km above sea level, every 2 km, <http://puff.images.alaska.edu>. These simulations are displayed online for ease of access and through auto-updating pages. This allows the analyst to link to one internet location to view the most recent VATD simulation. The Puff model simulations are used by the Alaska Volcano Observatory, U. S. National Weather Service, and numerous other agencies worldwide to assist in volcanic ash cloud hazard assessment. Simulations of the ash cloud for up to 24 hours in the future and hourly snapshots are provided. These are useful prior to eruptions as hazard assessment maps to predict potentially impacted areas. The model simulations are provided within a Virtual Globe format to analyze the three-dimensional structure of the ash cloud movement and to allow easy data fusion with other data sources. These datasets can be viewed as a ‘network link’, again allowing the analyst to view the most recent simulation from any location with internet access. Lagrangian models, such as Puff, can be run in minutes and therefore multiple scenarios can be developed without the need for high performance computing. Here, we show that knowledge of the pre-eruptive ash cloud hazard is critical to generate a quick, accurate assessment and there is a need, during a crisis, to be able to directly access real-time ash cloud simulation without the need to initialize the model and wait for results.

Webster, Helen N.

Forecasting Peak Ash Concentrations within the Volcanic Cloud from the 2010 Eruption of Eyjafjallajökull, Iceland

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Prolonged disruption to aviation during the April - May 2010 eruption of the Icelandic volcano Eyjafjallajökull resulted in pressure to estimate ash concentrations within the predicted volcanic cloud for the purpose of allowing aircraft to fly in regions with low ash contamination. As a result the modelling procedure at the London VAAC (Volcanic Ash Advisory Centre), using the Lagrangian dispersion model NAME (Numerical Atmospheric-dispersion Modelling Environment), changed from forecasting solely regions of significant ash to also estimating peak ash concentrations. This required consideration of estimates of the volcanic ash emission rates,

near-source fall-out rates of large ash particles and of aggregates of ash particles, and the relationship between predicted mean ash concentrations and localised peak concentrations unresolved by the model. The model set-up used operationally towards the end of the 2010 Eyjafjallajökull eruption to forecast peak concentrations within the ash cloud is described together with different model set-ups subsequently investigated and based on predicting concentrations within thinner model layers. The performance of NAME (the operational model of the London VAAC) in predicting peak ash concentrations observed during the eruption is assessed. The agreement between model predicted and observed peak ash concentrations is in line with what is expected for a dispersion problem of this type where there are many sources of error. The results are comparable for the different model set-ups with modelled values within about an order of magnitude or so of observations, in most case. Introduction of a buffer zone, to account for small positional errors in the predicted plume location, is found to significantly improve the agreement between model predictions and observations. The sensitivity of model predictions to the source detail, in particular, the eruption height and the vertical distribution of ash, is assessed. Errors in the source eruption height are found to be a significant contributor to errors in the predicted ash concentrations.

Wernli, Heini

The sensitivity of offline trajectory calculations on the temporal resolution of the meteorological input data

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Most trajectory studies are performed offline (or “post mortem”), using wind fields (and other meteorological parameters) available at distinct time steps from (re)analyses or numerical model output. Typically, for (re)analyses the temporal resolution of the input data is six hours. The time step used for calculating trajectories is much smaller and therefore the offline computation of trajectories is only possible when interpolating the available wind fields in time (and space). This temporal interpolation issue constitutes a major limitation of the accuracy of trajectories, in particular in regions characterized by a large spatio-temporal variability of the vertical wind field (e.g., near fronts and in regions of deep convection). In this study, a sensitivity experiment is performed using hourly meteorological fields from a special 5-day forecast of the global ECMWF model with a spectral resolution of T799L91. Trajectory calculations are intercompared when using 1, 2, 3, and 6 hourly input fields. The evaluation is performed both statistically and with a dedicated focus on key airflows in extratropical weather systems. The statistical results, using previously suggested

measures for deviations between trajectories, the “absolute horizontal/vertical transport deviation” AHTD and AVTD, respectively, indicate a rapid separation of trajectories starting from the same grid points when using input fields with different temporal resolution. For instance the AHTD, averaged in the Northern Hemisphere, grows to 250 km within 5 days when comparing trajectories with 1 and 2 hours temporal resolution. The comparison for resolutions of 1 and 6 hours leads to an average increase of AHTD to 800 km within 5 days. This indicates a substantial source of errors for trajectories computed with the standard temporal resolution of (re)analysis data of 6 hours. In a second part of the study, we investigate the effect of the temporal resolution of the input data on the identification of key flow features, including warm conveyor belts, dry descending airstreams, tropical moisture export events, and stratospheric intrusions. The sensitivity of the frequency of these events to the temporal resolution and of their overall physical characteristics (e.g., overall diabatic heating rate within warm conveyor belts and potential vorticity conservation along stratospheric intrusions) will provide important information about the accuracy of these features identified from routinely available (re)analysis data sets.

Wernli, Heini

A Lagrangian method to identify regions potentially affected by wet deposition of radioactive emissions from the Fukushima power plant - a climatological study

Wernli, Heini¹; Sodemann, Harald¹

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The earthquake and tsunami that affected Northern Japan on 11 March 2011 produced severe damage at the Fukushima nuclear power plant. Radioactive emissions occurred during the following days and weeks, both directly into the atmosphere and the ocean. However, the temporal evolution of the amount and type of emissions remains unknown. The immediate and most severe implications are mainly occurring in the direct neighborhood of the Fukushima power plant. Remote effects of the radioactive emissions can occur, for instance, if the emitted substances are transported in the atmosphere and deposited at the surface by dry and wet deposition. In principle, wet deposition of radioactive substances can occur at distances of several 1000 km from the source. However, emissions can only reach remote areas if they are not totally washed out along their pathway to the remote area. It is the aim of this study to identify the geographical regions that can be potentially affected by wet deposition of radioactive emissions from the Fukushima power plant. To this end a high-quality global meteorological data set for the years 1989-2010 (ERAinterim) and a relatively simple Lagrangian approach are used for identifying potential wet deposition areas of Fukushima emissions in a climatological sense. Such a climatological analysis enables us to address the following questions: (1) Which areas are extremely unlikely affected by wet deposition of emissions from the Fukushima

power plant?, (2) Which areas (in addition to Japan) are potentially most endangered by wet deposition of Fukushima emissions?, and (3) How do these areas change from spring to summer (March until June) and why? It is shown that the potentially most affected region is the Pacific basin, with remarkable intra-seasonal variations. In spring, in addition to the main transport pathway towards the east, the preferred occurrence of surface high-pressure systems to the south of Japan leads to relatively strong transport towards the Philippines. However, in June the key region of high-pressure systems shifts to the north of Japan associated with a westward transport pathway from Japan into the Asian continent.

White, Alex

Source sensitivity in surface-to-stratosphere transport addressed using an Eulerian and a Lagrangian adjoint model

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2. Department of Chemistry, University of Cambridge, Cambridge, United Kingdom

Adjoint chemistry transport models are increasingly being exploited to ask fundamental questions about the nature of atmospheric transport. For example, the question of how the efficiency of surface-to-stratosphere transport (SST) depends upon source location, is best addressed using an adjoint model. One particular formulation of the adjoint problem, leading to the retro-transport equation of Hourdin and Talagrand (QJRM, 2006), is the natural one to address a given problem using both Eulerian and stochastic Lagrangian methods. Here, the dependence of SST upon source location is studied using both a retro-transport version of the Eulerian model TOMCAT, and the Lagrangian particle dispersion model FLEXPART in adjoint (back trajectory) mode. The results from each type of model can be presented in the form of sensitivity maps showing the relative likelihood of SST from different source locations. Both models confirm that very-short-lived species are much more likely to reach the stratosphere if they are emitted from certain ‘hot-spot’ locations. Issues arising from cross-validation between the two models are discussed.

Wilson, John D.

Are “Rogue Velocities” in Lagrangian Stochastic Simulations Other Than a Manifestation of an Insufficiently Small Time Step?

Wilson, John D.¹

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Several people have reported the experience of a well-mixed Lagrangian stochastic model generating unrealistic velocities, and ad hoc steps have been taken to control the impact of this phenomenon. The occurrence of these “rogue velocities” would on first sight appear to contradict the model’s being well-mixed, however the problem is typically

experienced in the context of a complex (and in some cases, discontinuous, i.e. gridded) regime of turbulence, corresponding to which there may (implicitly) be severe limitations on the allowable size of the time step - limitations that may not have been respected. This talk will summarize a playful look at this problem in an idealized regime of flow. An artificial regime of one-dimensional turbulence is defined, in which two regions of differing constant velocity variance σ_u^2 are joined by a ramp such that $\partial\sigma_u^2/\partial x$ is discontinuous at the ends $x=\pm D/2$ of the inhomogeneous region. The evolution of an initially well-mixed distribution of tracer is computed by integrating the Chapman-Kolmogorov equation, and it is found that tracer remains well-mixed provided that $\sigma_u \Delta t \ll D$.

Wilson, John D.

History of the Lagrangian Stochastic Model for Turbulent Dispersion, Part 1

Wilson, John D.¹; Sawford, Brian²; Thomson, David³

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2. Mechanical and Aerospace Engineering, Monash University, Melbourne, VIC, Australia
3. Met Office, Exeter, United Kingdom

Some fifty years elapsed between Taylor's (1921) pioneering Lagrangian treatment of turbulent convection, and the implementation of Lagrangian models of dispersion in real (inhomogeneous) atmospheric flows. During that long interval the advantages of the Lagrangian viewpoint (among them, correct treatment of the non-diffusive "near field" of a source) were widely appreciated, but treatment of practically interesting flows hinged on the development of (and widespread access to) computers. This talk will survey early variants of the Lagrangian stochastic model, covering the period from Taylor (1921) until the early 1980's - by which time it had been shown that good agreement with observed dispersion in the simplest regime of inhomogeneous atmospheric turbulence could easily and rationally be procured, but that existing methods were unable to properly handle more general cases in which the turbulent velocity scale varies with position (as, for instance, within a forest canopy, or the convective boundary layer). Emphasis will be placed on the motivation and context of these early contributions. (Part 2 will cover the contributions that constrained the Lagrangian model in such a way as to largely solve this problem, cover outstanding difficulties and/or subtleties, and indicate the fascinating scope of subsequent applications of Lagrangian models).

Wilson, John D.

Inverse Dispersion in Disturbed Surface Layer Flows Using a Three-Dimensional Backward Lagrangian Stochastic ('3D-bLS) Model (*INVITED*)

Wilson, John D.¹; Flesch, Thomas¹; Bourdin, Patrick²

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2. Flight Sciences Group, Bombardier Aerospace, Toronto, ON, Canada

The "bLS" (backward Lagrangian stochastic) method for inverse dispersion most commonly treats the flow in which sources and detectors are embedded as being horizontally-homogeneous, and in the case that trajectories are confined to the surface layer needed wind statistics are provided by the Monin-Obukhov similarity theory - in which case the methodology can be titled "MO-bLS." Typically in practice, however, obstacles disturb the wind such that the flow is NOT truly horizontally homogeneous. At the cost of considerable trouble the bLS approach may be generalized for a refined treatment of such situations, it being one of the strengths of a well-mixed Lagrangian stochastic model that it is able to rationally incorporate all available statistical information on the flow (information that may be observational, or may stem from a computation). This talk will cover the authors' experience comparing "MO-bLS" and "3D-bLS" as applied to (inverse) dispersion from a small artificial area source of methane enclosed within a porous windbreak. This test case suggests that bLS (in general) is rather robust: source strength Q deduced from measured line-average concentration is found to be correct to within a factor of two, even if the (severe) flow disturbance is neglected. The more detailed 3D-bLS approach, driven by a RANS (Reynolds-averaged Navier-Stokes) computation of the horizontal gradients in wind statistics, results in a better estimation of source strength if the detector stands within the region of highly disturbed flow.

Wilson, John D.

Application of the Backward Lagrangian Stochastic (bLS) Method to Quantify Methane Emission from a Lagoon

Wilson, John D.¹; Flesch, Thomas¹; Mahzabin, Tarana¹

1. Earth & Atmospheric Sciences, University of Alberta, Edmonton, AB, Canada

In the context of trace gas dispersion, a backward Lagrangian stochastic model is a particularly convenient means to determine the relationship $n=UC/Q$ between the strength Q of an area source (of known perimeter) and the concentration C at a downwind point, for a given state of the atmosphere (reference wind speed U , etc.). The link between C and Q hinges on the distribution of touchdown points (where trajectories from the detector touch the ground within the source perimeter) and touchdown vertical velocities which, once computed, allow Q to be determined by measuring C . This talk will illustrate the eminent

convenience and practicality of the bLS approach, taking as an example recent measurements of methane emission from a municipal waste lagoon, a case which will illustrate the flexibility of the method and the key role of user judgment and prior information.

Wohltmann, Ingo

The Lagrangian Chemistry and Transport Model ATLAS

Wohltmann, Ingo¹; Rex, Markus¹; Lehmann, Ralph¹

1. Alfred Wegener Institute, Potsdam, Germany

ATLAS is a global Chemistry and Transport Model (CTM) based on a Lagrangian approach with a focus on the stratosphere. We give an overview over the model implementation and show some selected applications of the model, including e.g. simulations of ozone depletion and denitrification in the Arctic, estimation of diffusion coefficients in the stratosphere, determining the latitudinal and vertical origin of air masses enclosed in the polar vortex or transport of short-lived bromine species. Transport and chemistry in the model are typically driven by ECMWF reanalysis data. With this input, a large number of trajectories is initialized and advected, filling the domain of the complete atmosphere. Chemistry is calculated on every trajectory like in a box model. An important feature of the model is the mixing algorithm, which simulates atmospheric mixing and diffusion based on a physical approach using properties of the wind fields (i.e. shear and strain) in a way similar to the CLaMS model. This approach is not only necessary in Lagrangian models (which show no numerical diffusion like Eulerian models), but also allows for a realistic simulation of atmospheric diffusion by tuning the algorithm to observations and keeps the density of air parcels constant and homogeneous. The model includes a stratospheric chemistry module, heterogeneous chemistry on polar stratospheric clouds and a particle-based Lagrangian denitrification module. The chemistry module comprises 46 active species and 171 reactions.

Heterogeneous chemistry is based on the equilibrium module of Carslaw and simulates several reactions on polar stratospheric clouds. The denitrification module simulates the nucleation, transport, sedimentation and growth of a large number of polar stratospheric cloud particles and is based on the DLAPSE model.

Wotawa, Gerhard

Simulation of the transport of radioactivity from the Fukushima nuclear accident

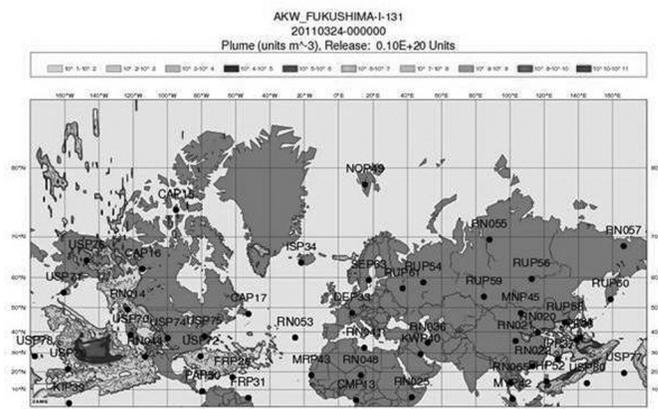
Wotawa, Gerhard¹

1. Data, Methods and Modelling, Central Institute for Meteorology and Geodynamics, Vienna, Austria

On 12 March 2011 at about 6:30 UTC, the first explosion was reported from block 1 of the nuclear power plant in Fukushima Daiichi. Only minutes afterwards, the Meteorological Service of Austria (ZAMG) started its model simulation of the event. As transport model, the Lagrangian

Particle Diffusion model FLEXPART Version 8 based on ECMWF input data was used. The simulated substances were I-131, Cs-137 and Xe-133. The simulation showed a hemispheric-scale spread of radioactivity during the first 14 days (see Figure 1). ZAMG has real-time access to the global radioactivity data of the CTBT Organization (CTBTO). CTBTO stations are distributed all over the globe and measure radioactive particles as well as Xenon gases with very high accuracy. These data were used to validate the model simulation, and to estimate the source terms of I-131, Cs-137 and Xe-133. First results show that the model worked well in predicting the radioactivity detections in North America as well as Europe (see Figure 2). The concentrations of I-131 transported towards Europe were underestimated. This was due to the fact that a significant percentage of I-131 was released and transported as gas. Regarding emissions from Fukushima, the comparison of model results with measurements demonstrated that significant amounts of I-131, Cs-137 and Xe-133 were set free during the first days. The estimates for I-131 amounted to 10^{17} Bq, for Cs-137 to 10^{16} Bq per day. This is on the same order of magnitude as daily emissions of these substances from the Chernobyl accident. Our estimates exceeded initial estimates from Japanese authorities, IAEA and CTBTO by orders of magnitude.

<http://www.zamg.ac.at>



Zeng, Jiye

Introducing an Implementation of Parallel Computing for Lagrangian Modelling of Particle Dispersion in the Atmosphere

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Lagrangian particle dispersion models are often used to study the transport and diffusion of tracers in the atmosphere. A typical application requires a large number of particles when they eventually spread to a wide area. As a result, computing the position changes of particles becomes the bottleneck that controls the total execution time of such models. We introduce an open-source freeware that implements parallel computing to accelerate Lagrangian modelling of particle dispersion in the atmosphere. We explored the parallel computing potential of the NVIDIA's Graphic Processor Unit (GPU) and its limitations for our application. Our experiments using a 8-core CPU with a Tesla C2050 GPU showed that the performance gain of parallelism over non-parallelism could be as high as 20 times. Being able to produce modelling results quickly is important in responding to such a disaster as the radiation leak from the Fukushima Daini Nuclear Power Plant of Japan, which was caused by the earthquake and tsunami on 11 March 2011. We used the observed radiation dose rate at Tsukuba, Ibaraki, Japan, to verify the model by releasing 50,000 particles to the location of the power plant at time interval of 3 hours and simulating the particle dispersion for 10 days. A simulation took less than 5 minutes by a PC with a 4-core CPU of 2.00GHz and a Tesla C1060 GPU. The first two waves of particle arrival on March 15 and 21, indicated by long particle residence times, agree well with the observed surges of the dose rate (Figure 1). In other periods of long residence times after March 28, the dose rate did not show significant increases, indicating that after the explosions in the early days at the power plant, the radiation release to the atmosphere had been stable, even if not under control.

<http://db.cger.nies.go.jp/metex/flexcpp.html>

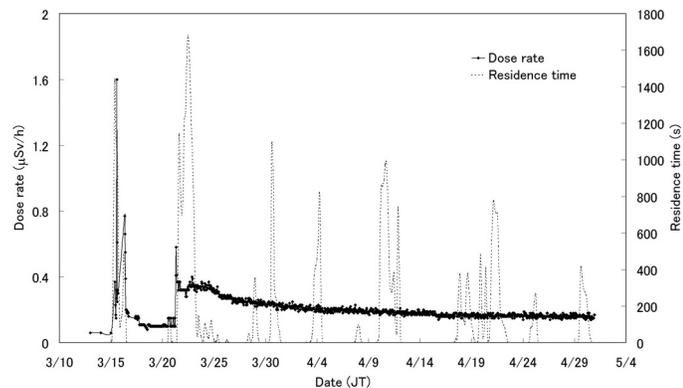


Figure 1. Observed radiation dose rate and modeled residence time. The National Institute of Advanced Industrial Science and Technology made the observation and disseminates data at http://www.aist.go.jp/taisaku/ja/measurement/all_results.html. The end time of background radiation was estimated based on the information at <http://www.kek.jp/quake/radmonitor/>. The residence time is the mean of all 0.25x0.25 grids in a 1x1 grid area around the Tsukuba Center.