THE ROLE OF ATMOSPHERIC BOUNDARY LAYER PROCESSES IN ATMOSPHERIC CHEMISTRY

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The importance of atmospheric boundary layer (ABL) dynamics and physics in controlling key aspects of atmospheric chemistry is becoming increasingly recognized. Processes such as dispersion, mixing, transformation, and deposition are strongly dependent on the stratification of the ABL, on the presence of clouds, and on the interaction between land and the ABL. Its location as a buffer region between the biosphere and the free troposphere makes this layer very relevant to air quality issues and the global climate problem. Therefore, to obtain a thorough understanding of the temporal evolution and spatial distribution of atmospheric compounds, it is necessary to investigate the myriad linkages between the prevailing physical processes and the chemistry of their constituents.

In the past, these subjects have often been investigated separately, eliminating the possibility for study of the potential feedbacks and couplings of the relevant processes in atmospheric chemistry. Toward the purpose of rectifying this historic segregation, the interdisciplinary "Research School" was organized to narrow the gap between the disciplines of atmospheric physics and chemistry, identify significant gaps of knowledge, and promote vital cross-disciplinary dialog among emerging scientists in both fields.

In November 2006, we held a 5-day Research School attended by 27 students from 12 different

RESEARCH SCHOOL ON THE DISCIPLINES OF ATMOSPHERIC PHYSICS AND CHEMISTRY

WHAT: Junior and senior researchers identified, discussed, and studied outstanding issues related to atmospheric boundary layer processes closely linked to atmospheric chemistry
WHEN: 20–24 November 2006
WHERE: Castelldefels, Spain

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nations with different backgrounds in atmospheric chemistry and physics. Nine lecturers explained the fundamental concepts, provided an overview of the current understanding of the subjects, and attempted to identify future research issues. The lectures were complemented by four afternoon practical exercises to apply some of the concepts explained during the morning lectures. The last day was devoted to oral presentations given by the participants. The combination of oral lectures given by senior researchers with the practical exercises proved to be an excellent format to create a discussion forum and learn about possible links between the fields. This was reflected in the discussion on the last day during which the junior scientists proposed challenging research to be tackled in future investigations.

Because of the large span of spatial and temporal scales that characterize the processes in both fields, special care was taken to maintain a good balance of subjects and stress interdisciplinary aspects. At the smaller scales, emphasis was placed on microphysical processes that lead to different chemical paths and aerosol formation. Small-scale turbulence might also enhance or limit the ability of cloud droplets, aerosols, and reactive species to efficiently engage in processes such as coagulation, coalescence, chemical transformation, and deposition. Moving to large scales, the issue of boundary layer clouds was addressed in the Research School because cloud formation and cloud structure are closely linked to the above-mentioned small-scale processes. In addition, clouds modify the structure of the boundary layer by enhancing the vertical transport of atmospheric compounds. Atmospheric stability can inhibit cloud formation, but can also either largely reduce or enhance the dispersion and mixing of species. All these processes and phenomena are controlled by the large diurnal variability over land and the large modifications of the boundary layer depth related to turbulence intensity. In addition, boundary layer development can be strongly perturbed by the presence of a canopy, topography, or induced mesoscale atmospheric circulations caused by surface variations, such as roughness length and soil moisture.

LECTURES. Jordi Vilà-Guerau de Arellano (see title page for affiliations) opened the Research School with a talk on the relevant aspects that influence atmospheric chemistry under diurnal convective conditions. Near the surface, the reactivity of species and aerosols can lead to deviations from the "classical" concept of constant flux in the atmospheric surface layer, implying that the deposition velocities of the atmospheric compounds vary with height. He then discussed the role of large eddies in controlling the chemical reactivity and the additional segregation of species caused by the nonuniformity of the emissions. Presenting large-eddy simulations of turbulent reacting flows in idealized boundary layer conditions, he showed that turbulent fluctuations that create the inefficient mixing can perturb the chemical equilibrium and slow down the chemical transformations, relative to the expectation of a perfectly well-mixed reactor. In spite of the development of new instruments, the lack of experimental evidence is, to a certain extent, hindering the possibility of advancing this theory. His talk concluded with an illustration of the importance of entrainment as the process at the top of the ABL that allows the exchange of atmospheric compounds between the boundary layer and the free troposphere.

The roles of stratification and surface characteristics were recurring themes in the presentation on nocturnal chemistry by Jochen Stutz, and on the role of the canopy and marine environments in chemistry by Ian Faloona. Stutz said there is now convincing experimental evidence that stable stratification is a key factor in the vertical distribution of ozone at night. Based on a combination of observations and modeling, he showed that an altitude-dependent chemistry in the stable nocturnal boundary layer (NBL) leads to zones where different chemical mechanisms deplete ozone and other pollutants. Near the ground chemistry is impacted by the upward transport of freshly emitted species, while the top of the NBL is influenced by the entrainment of compounds from the residual layer. A reactive zone develops between these two borders where active radical chemistry occurs. As a result of emissions, transport, and entrainment, the reaction zone evolves in time from being near the surface (close to the sources) to the top of the NBL. Turbulent intermittent events can mitigate this layering by transporting and mixing species from adjacent levels.

The interaction of layers with differing static stability, such as those encountered in natural plant or urban canopies, made Faloona stress the importance of mixing near the emission and deposition sources and sinks, and in particular the dire inadequacy of eddy diffusivity models in situations where the turbulent mixing elements were of the same size or larger than the principal features of the concentration gradients. Under such flow conditions, it can be quite useful to represent the intermittent ejections and sweep events that characterize turbulence within canopies, because these features can be formally analyzed to estimate mean fluxes using a surface renewal model. He concluded his lecture by discussing the role of marine boundary layers in atmospheric chemistry, and some of the most stark differences from their textbook continental counterparts. He also emphasized the influence of biological processes on atmospheric chemistry and physics through the relation of dimethylsulfide emissions to the formation of cloud condensation nuclei and the evolution of marine boundary layer clouds.

Both Stutz and Faloona described the development and application of new instruments that can help improve our understanding of atmospheric compounds in the boundary layer and at the surface. The differential optical absorption spectroscopy (DOAS) has proven to be very efficient at measuring vertical profiles of atmospheric compounds continuously in urban regions. Other instruments based on fast analytical techniques, such as proton transfer reaction mass spectrometry (PTR-MS) or chemiluminescence, in combination with standard micrometeorological techniques, such as ultrasonic anemometry, currently open up the possibility of measuring fluxes of reactive atmospheric compounds, which was previously not possible, using the eddy covariance technique.

Stefano Galmarini surveyed the role of atmospheric turbulence in dispersing atmospheric compounds. This lecture concentrated on point releases represented as a paradigm of any other kind of dispersion (e.g., line, area). After a short historical overview the students were asked for an interpretation of several images of dispersing clouds and to work out the probable phenomenology suggested by the cloud morphology. The common problem of determining dispersion parameters presented as the synthesis of turbulent processes was then tackled from the early heuristic formulations to those based on the latest results using the large-eddy simulation technique. Plume rise and removal mechanisms followed with special emphasis on chemical reaction and the role of turbulent mixing in controlling the reactivity in a dispersing cloud. The more advanced approaches of Eulerian and Lagrangian dispersion models were then presented as suitable approaches for simulating the short, medium, and long ranges. Galmarini concluded by presenting examples of future research issues, such as the dispersion in urban areas, which is usually characterized by the heterogeneity of the emissions.

The role of sea-breeze circulations, flows driven by orography, and thermal lows on air pollution were thoroughly discussed by Millán Millán. These mesoscale flows strongly perturb the boundary layer development, and, as a consequence, the distribution of greenhouse gases and reactive compounds that are emitted or formed are mainly driven by these mesoscale circulations. The budget of these atmospheric compounds is largely controlled by the coupling of surface, boundary layer, and mesoscale circulations during the day. Large amounts of the compounds are transported to the sea by return circulations aloft, and are stored in layers above the sea during the night. The onset of diurnal processes transports them back to the land. Millán closed his talk by pointing out the modeling challenges ahead, particularly in meteorological situations characterized by the relation between boundary layer processes and induced mesoscale phenomena.

Progress and challenges in understanding the feedback between cloud dynamics, microphysics, and chemistry were the topics of several lectures. Harm Jonker reviewed the main concepts of wet thermodynamics and the implications in the description of boundary layer clouds. The presence of clouds, for instance, fair weather clouds or shallow cumuli, modifies the structure and growth of the boundary layer, promoting the formation of thermal chimneys that enhance the vertical transport of pollutants emitted at the surface. He stressed the need to thoroughly understand the dynamics that relate the subcloud layer and the cloud layer to the surrounding environment in order to assess the fate of the atmospheric compounds. Jonker further explained that the characterization of these cloudy boundary layers is linked to perturbations of the chemical equilibrium caused by the clouds themselves disturbing the ultraviolet radiative transfer.

Harry ten Brink reviewed the role of aerosols in cloud formation and chemistry, and their role, not yet well understood or quantified, in regulating the climate. He pointed out some of the difficulties in measuring them because of the semivolatility of many of their components. Based on continuous observations of ammonium nitrate at Cabauw, Netherlands, he showed the need to analyze the measurements together with observations of surface processes and boundary layer dynamics.

Nadine Chaumerliac discussed the role of cloud microphysics and dynamics. She emphasized the importance of maintaining a balance in parameterizations of clouds to include radiation processes (at the mesoscale), dynamics (cloud scale), microphysics (particle scale), and multiphase chemistry (at the smallest molecular scale). The aerosol and cloud microphysics, and in particular the interactions between aerosol particles increasing with human activity and water and ice particles, affect the fate of chemical species dissolved in the drops. Using a different approach than Jonker, Chaumerliac concluded that there remains a need to study the coupling of boundary layer clouds and convective clouds in venting pollutants to the free troposphere.

The oral presentations on the final day, which also stressed the interdisciplinary aspects, completed the lecture themes (for more information see the Research School Web page at autumnschool.ablresearch.org).

PUTTING WORDS INTO PRACTICE. Four practical exercises inspired by current research model activities or field experiments gave the participants a chance to become acquainted with current research tools related to the morning lectures. The four practical exercises focused on the following:

a) the carbon dioxide budget in the atmospheric convective boundary layer (using a mixed-layer model);
b) entrainment, convection, and deposition in the stratocumulus-topped marine boundary layer [analyzing and treating observational data from the Second Dynamics and Chemistry of Marine Stratocumulus (DYCOMS-II) experiment];
c) cloud dynamics and transport of reactive species (using results from large-eddy simulation numerical experiments); and
d) cloud chemistry (using a box model).

A debate to explore future lines of research and collaboration closed the Research School. The participants were divided into three groups and asked to formulate a scientific proposal. In true brainstorming fashion, the three groups discussed and debated, while attempting to identify potential gaps of knowledge in the fields of atmospheric chemistry and physics. Despite the relatively short time frame and the spontaneity of the event, the following three research proposals were formulated:

a) define and establish a coordinated database of aerosol observations that is efficiently related to model activities;
b) improve knowledge of the processes necessary to exchange atmospheric compounds between the ABL and the free troposphere; and
c) identify and understand the physical/chemical problems of so-called "megacities" that are rapidly developing in the twenty-first century.

Representatives of the three groups presented their proposals and actively defended them during the ensuing plenary discussion in which lecturers and members of the other groups asked questions and offered critical comments. Resulting discussions centered on four points: originality, relevance, feasibility, and interdisciplinarity. For the sake of synthesis, we will focus only on the third project, because it reflects the spirit of the Research School so well and, to some extent, encompasses the other two research proposals.

The increase of the urban region dimensions of the cities planned for the twenty-first century represents a challenge for the atmospheric physics/chemistry community because it requires maintaining air quality standards in relation to both human health and the potential impact on climate change. The importance of having a reliable database of emissions (aerosols and atmospheric compounds) as mentioned in project (a) is crucial to modeling future air pollution scenarios accurately and establishing effective mitigation policies. However, this must be done simultaneously with a thorough understanding of the most relevant physical/chemical processes [such as the one described in project (b)]. For example, important atmospheric phenomena, such as clouds and their role in the transport of air pollutants, can be largely dependent on freshly emitted aerosols, on induced turbulence by urban canopies, and on atmospheric flows driven by surface heterogeneities.

In conclusion, a reliable representation of the future impact of megacities in the air quality and climate problem can only be realized if we are able to obtain a balance in the complexity of processes and disciplines. In pursing this objective, this Research School represented an attempt to bring together scientists with different experience and backgrounds and to establish closer links between the atmospheric physics and chemistry communities.

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