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Overview of the Special Issue "Selected Papers from the 2nd Atmospheric Chemistry and Physics at Mountain Sites Symposium"

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ABSTRACT

Mountain sites provide a unique window on atmospheric chemistry and physics. These sites allow for continuous observations at high elevation, often in the free troposphere, where many important processes occur. Observations at these mountain sites allow for studies on long-range transport of pollution, cloud and precipitation processes, boundary layer ventilation and long-term observations of climate relevant gases and aerosols. However operating at mountain sites presents a unique set of challenges, and for this reason scientists doing research at these sites sought a forum to share knowledge on both the science and challenges of working at these sites.

THE ATMOSPHERIC CHEMISTRY AND PHYSICS AT MOUNTAIN SITES SYMPOSIUM SERIES.

The first Atmospheric Chemistry and Physics at Mountain Sites Symposium was held June 8–10, 2010 at Interlaken, Switzerland. The second Symposium was held Aug 11–15, 2014 in Steamboat Springs, CO, USA. This 2nd symposium provided a platform to present and discuss observations of air constituents at mountain sites.

The goal of these meetings is to learn as much as possible from mountain measurements across the globe. Since many of the scientists representing mountain sites have similar types of scientific questions and challenges this symposium enabled fruitful scientific discussions and collaborations. Sessions consisted of relatively short presentations on August 12–14 in order to accommodate open discussion periods. An optional tour of Storm Peak Laboratory (www.stormpeak.dri.edu) took place on August 11 and August 15. Thirty-six presenting scientists attended the workshop representing twenty-three different institutions. The workshop consisted of 45 oral presentations spread across the following seven sessions:

Overview of Scientific Activities at Specific Mountain Sites;

- Biomass Burning;
- New Particle Formation;
- Trace Gases at Mountain Sites;
- Aerosol Properties at Mountain Sites;
- New Approaches at Mountain Sites; and
- Challenges and Lessons Learned working at Mountain Sites.

The following mountain research stations were represented at the symposium:

- Mt. Fuji Weather Station, Japan
- Mt. Bachelor Observatory, Oregon, USA
- Storm Peak Laboratory, Colorado, USA
- Whistler Peak, British Columbia, Canada
- Mt. Lulin, Taiwan
- Sonnblick Observatory, Austria
- Jungfraujoch High Altitude Research Station, Switzerland
- Mt. Izaña, Tenerife, Canary Islands, Spain
- Pico Mountain Observatory, Azores, Portugal
- Greenland Environmental Observatory (GEOSummit), Denmark
- Whiteface Mountain Adirondack High Peaks Observatory, New York, USA
- AppalAIR, NC, USA
- Pinnacles Natural Area Preserve, Virginia, USA
- Atmospheric Research Observatory at the Amundsen-Scott South Pole Station

Additionally, results from other mountain sites were also presented via collaboration.

The workshop proceedings are available here: http://stormpeak.dri.edu/images/stories/workshops-conferences/SPL_2014_Symposium_Program.pdf.

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In the Special Issue

The general aim of this special issue is to present results from field measurements of atmospheric constituents at mountain sites across the globe, including model evaluation of these measurements. Mountain sites are important points of observation because they are at the interface between the atmospheric boundary layer and free troposphere; when they are uninfluenced by local pollution sources, mountain sites represent the regional atmospheric background. In soliciting articles for this special issue, we encouraged papers presenting scientific findings from mountain sites focused on aerosol (chemical, physical, and optical properties), gas-phase (reactive short lived and climate-active long live trace gases), cloud (microphysical properties and chemistry), and/or solar radiation measurements. For example, mountain sites are often used to study the influence of local pollution, long-range transport or new particle formation. Second, we encouraged papers presenting global and regional model comparisons to mountain site measurements. Examples include the assessment of long-range transport in the models, and universal issues of comparing models to mountain measurements (e.g., complex terrain, upslope/downslope flow, potential for large sub-grid variability of atmospheric properties). Seasonal cycles due to changing sources (for instance, dust, biomass burning) at mountain sites are often seen (e.g., Andrews *et al.*, 2011), offering an opportunity/challenge for model comparisons. Finally, we encouraged papers focused on sampling techniques and data analysis that is unique to mountain sites (e.g., inlet designs, designing equipment to deal with riming conditions, methods for identifying in-cloud or free tropospheric conditions).

This special issue of the journal includes 34 papers selected from submissions from conference attendees as well as the extended mountain measurement community after a rigorous peer-review process. The final papers selected represent five general themes covering topics related to regional air pollution; the interaction between air and precipitation chemistry; the classification of and the exchange between the free troposphere and boundary layer; aerosol and trace gas climatology; and model evaluation.

These papers represent findings from 34 sites across six continents (Antarctica, Asia, Australia, Europe, North America and South America) as listed in Table 1 and illustrated in Fig. 1. Twenty-nine of these sites are high elevation locations, and five sites are either lower elevation sites in mountainous regions or low elevation sites with data used in direct comparison with mountain measurements.

Regional Air Pollution

Mountain sites have a unique vantage point for observing regional air pollution. Ten papers within this special issue focused on the observations of regional pollution from a high elevation. Different methodologies were used depending on the site and regional needs.

Forty-five ambient air samples collected in the summer of 2011 and spring of 2012 on Mount Lushan in Southern China, a mountain site surrounded with industrial districts. The samples were analyzed for aromatic hydrocarbons and halocarbons (Yang *et al.*, 2016). Backward trajectories of the sampled air masses and the corresponding daily concentrations of individual species suggest that long-range transport has a strong effect on the sampling site, and specific air masses were primarily influenced by automotive emissions. Kumar and Attri (2016) measured carbonaceous aerosol at a site in the western Himalayas and found that a significant fraction (26%) of the PM_{10} aerosol was carbonaceous, with biomass fuels the most important source. In another study from the Northwestern Himalayas (Himachal Pradesh, India), Saxena *et al.* (2016) performed laboratory-based emission factors and budget estimations for chemical species emitted from residential cooking-related biomass burning. They concluded that PM emission factors for dung cake burning are 3–4 times higher than residential wood combustion. Necki *et al.* (2016) report on observations of CH_4 and N_2O at a Polish station in the High Tatra Mountains and compare these data with CH_4 observations from the GOSAT satellite. One particularly interesting result is the findings of high CH_4 emissions from the Silesian Coal Basin and their influence on the station. Lo Vullo *et al.* (2016) report 5 years of observations of non-methane hydrocarbon observations from the Monte Cimone site in Italy. These authors report a significant decline in some hydrocarbons associated with reductions in regional emissions.

Cordova *et al.* (2016) employed field campaign measurements, models and trajectories to investigate under what synoptic conditions pollution might reach a mountain site in the Andes. They found that pollutant transport to the sampling site was enhanced on clear days relative to cloudy days for the time period of the study. A main impetus for Cordova *et al.* (2016) was to understand aerosol deposition on the snow – this also was the impetus for the work of Axson *et al.* (2016) who studied dust deposition on snow in the San Juan Mountains in Colorado. Axson *et al.* (2016) looked at both the chemical composition and size of deposited aerosol and using back trajectories were able to tie differences in observed composition to shifts in the mineral dust source region.

Galindo *et al.* (2016) studied the factors affecting the chemical composition of PM_1 at Mt. Aitana in Southeastern Spain. Organic matter and ammonium sulfate were the major contributors to PM_1 mass, while elemental carbon and nitrate, largely related to local sources, were minor components. Notably, nitrate concentrations were significantly lower than those measured at other elevated sites in Europe due to increased thermal decomposition of ammonium nitrate. In another case study at Sonnblick, Austria, Schauer *et al.* (2016) deployed aerosol absorption coefficient measurements as a tool to apportion biomass burning and Saharan dust in the air masses arriving at the sites, during an episode with increased PM levels.

The contribution of biomass burning from residential heating on ambient $PM_{2.5}$, especially prevalent in many mountain regions, is highlighted in Busby *et al.* (2016). This paper compares different complementary methods for apportionment of residential wood smoke, focusing on data from Fairbanks, Alaska. The results suggest that, collectively, chemical mass balance, levoglucosan analysis and ^{14}C analysis confirm apportionment results.

Table 1. Table of sites in Special Issue.

Location, country	Map ID	Coordinates lat. long.	Elevation (m asl)	Citation
Canaan Valley Institute, United States	CVI	+39.06 –79.42	988	Andronache, 2016
Cape Grim, Australia	CGO	–40.68 +144.68	160	Chambers *et al.*, 2016
Changbai Mtn.,China	CHM	+40.70 +127.70	2740	Lu *et al.*, 2016
Chilan Mtn, Taiwan	CHI	+24.58 +121.40	1650	Simon *et al.*, 2016
Dinghu Mtn., China	DIM	+23.15 +112.50	1000	Lu *et al.*, 2016
Fairbanks, United States	FAK	+64.84 –147.72	130	Busby *et al.*, 2016
Gulmarg, India	GUL	+34.06 +74.38	2690	Kumar *et al.*, 2016
Himachal Pradesh State, India	HIM	+31.10 +77.17	450–6800	Saxena *et al.*, 2016
Jungfraujoch, Switzerland	JFJ	+46.54 +7.98	3580	Bukowiecki *et al.*, 2016 Chambers *et al.*, 2016
Kangra, India	KAN	+76.3 +31.7	650	Kumar and Attri, 2016
Kasprowy Wierch, Poland	KWP	+49.23 +19.88	1989	Necki *et al.*, 2016
Kinmen Island, Taiwan	KII	+24.40 +118.30	48	Simon *et al.*, 2016
Mauna Loa, United States	MLO	+19.53 –155.57	3397	Sharma *et al.*, 2016 Chambers *et al.*, 2016
Monte Cimone	CMN	+44.20 +1070	2165	Lo Vullo *et al.*, 2016
Mount Aitana, Spain	MTA	+38.39 –0.27	1558	Galindo *et al.*, 2016
Mount Bachelor, United States	MBO	+43.98 –121.69	2763	McClure *et al.*, 2016
Mount Lulin, Taiwan	LLN	+23.47 +120.87	2862	Simon *et al.*, 2016
Mount Lushan, China	MTL	+29.57 +115.97	1165	Yang *et al.*, 2016
Mount Muztagh Ata, China	MMA	+38.28 +75.02	4500	Zhu *et al.*, 2016
Mount Tai, China	TAI	+36.26 +117.11	1500	Shen *et al.*, 2016
Mount Tateyama, Japan	MTJ	+36.58 +137.36	2450	Mochizuki *et al.*, 2016
Nam Co, China	NAM	+30.77 +90.99	4730	Kang *et al.*, 2016
Pico Este, United States	PRE	+18.27 –65.75	1051	Valle-Díaz *et al.*, 2016 Raga *et al.*, 2016
Puy de Dôme, France	PUY	+45.46 +2.57	1465	Barbet *et al.*, 2016 Freney *et al.*, 2016
Queltehues, Chile	QUE	–33.82 –70.22	1498	Cordova *et al.*, 2016
Sonnblick/Zittelhaus, Austria	SON	+47.05 +12.95	3106	Schauer *et al.*, 2016 Kirchner *et al.*, 2016
South Pole, Antarctica	SPO	–90.00 0.00	2835	Sheridan *et al.*, 2016
Storm Peak, United States	SPL	–84.99 –102.00	3220	Hallar *et al.*, 2016 Yu *et al.*, 2016
Swamp Angel Study Plot, United States	SAS	+37.91 –107.71	3371	Axson *et al.*, 2016
Weißfluhjoch, Switzerland	WEI	+46.83 +9.81	2663	Kirchner *et al.*, 2016
Whiteface Mountain, United States	WFM	+44.39 –73.86	1483	Brandt *et al.*, 2016 Schwab *et al.*, 2016a Schwab *et al.*, 2016b
Wuling Mountain, China	WUM	+40.38 +117.57	900	Du *et al.*, 2016
Xitou Experimental Forest, Taiwan	XEF	+23.65 +120.78	1150	Simon *et al.*, 2016
Zugspitze (Schneefernerhaus), Germany	UFS	+47.42 +10.98	2650	Kirchner *et al.*, 2016 Leuchner *et al.*, 2016

Interaction between Air and Precipitation Chemistry

Seven papers within this special issue focus on the interaction between air and precipitation chemistry at mountain sites, an important topic as mountains provide freshwater to half of the world's population (Kohler and Maselli, 2009).

Several studies within this special issue clearly demonstrated the impact of anthropogenic pollution on acidity of precipitation in mountainous regions. Kumar *et al.* (2016) reports snow chemistry and long-range transport of pollutants at Gulmarg in the northwestern Himalayan region of India during winters of 2012–2013. Data from this study were compared with a study from three decades ago, and a significant increase in the concentrations of anthropogenic components such as non-seasalt sulfate (114%), nitrate (109%) and ammonium (90%) was observed. The authors suggest this change was most likely due to increase in both long-range transport of pollutants as well as local activities during past three decades. Simon *et al.* (2016) characterizes and compares the chemical composition of fog water at four sites in Taiwan, of various altitudes. Coal combustion in Mainland China and Taiwan as well as

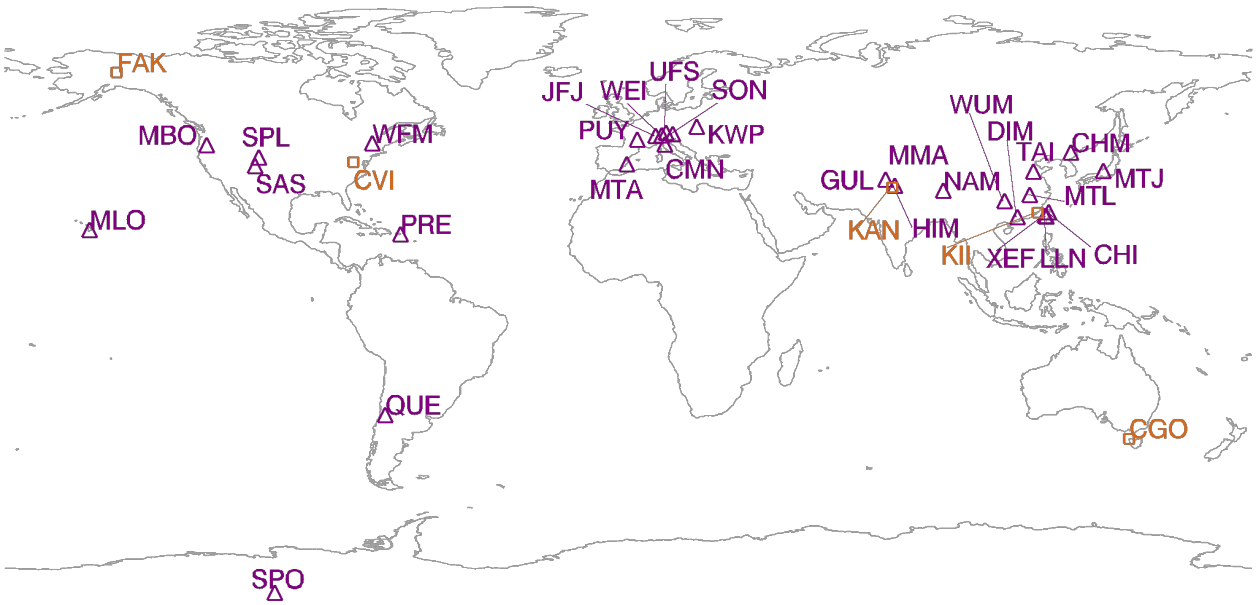

Fig. 1. Location of sites in this Special Issue. Sites in purple (triangles) are high elevation sites, sites in orange (squares) are either lower elevation sites in mountainous regions or low elevation sites with data used for comparison with mountain measurements.

nitrogen oxide emissions from urbanized central west Taiwan and the greater Taipei region were the main precursors of fog acidity. Andronache (2016) evaluated seasonal changes in insoluble ions in precipitation at a site in the Appalachian Mountains on the east coast of the United States. He found that there were distinct seasonal patterns with both greater precipitation and wet deposition of sulfate and ammonium ions in the summer. In contrast wet deposition of nitrate and marine related ions (sodium and chlorine) peaked in the winter, although precipitation was much less.

Lu *et al.* (2016) collected rainwater samples from forest ecosystems located in the Changbai, Wuling, and Dinghu mountains in eastern China to determine the microbial community compositions. Their results suggest that bacterial ice nuclei are present in the rainwater but play a minor role in ice nucleation in this region. Mochizuki *et al.* (2016) measured water-soluble organic nitrogen (WSON) and carbon in snow from a high elevation site in Japan. They find that WSON is closely connected to dust and pollution transported from the Asian continent.

At Pico Este in Northeastern Puerto Rico, bulk and drop size fractionated cloud water samples were collected at a Caribbean tropical montane cloud forest (Valle-Díaz *et al.*, 2016). One of their main findings was that long-range transported African dust impacts cloud water chemistry at Pico Este and that primary aerosols (i.e., mineral dust and sea-salt) were enriched in large cloud water droplets and secondary aerosols (i.e., anthropogenic particles) were enriched in small cloud water droplets. In another study from the same site, Raga *et al.* (2016) confirm, by the use of a novel depolarization mass spectrometer, that aged African dust particles are very good cloud condensation nuclei, while precipitation along air mass trajectories removes many particles.

Classification of and the Exchange between the Free Troposphere and Boundary Layer

Sampling techniques and data analysis for identifying free tropospheric conditions is a unique challenge to mountain sites, and four papers within this special issue focused on this topic. It is critical to distinguish between the planetary boundary layer and the free troposphere; as once aerosols or gases are transported into the free troposphere, their atmospheric lifetime can change significantly. Also trace gases and aerosols within the free troposphere tend to be representative of large spatial areas. McClure *et al.* (2016) examined data from the Mt. Bachelor Observatory in central Oregon to understand variations in carbon dioxide (CO_2) in the free troposphere and boundary layer. Free tropospheric air had a higher average CO_2 mixing ratios compared to the boundary layer air. This analysis also identified CO_2 events due to wildfires, the role of transport patterns, and the variation of CO_2 seasonally.

Evidence of anthropogenic emissions transported from the planetary mixing layer to the free troposphere was presented by Freney *et al.* (2016) using in-situ measurements of aerosol chemical and physical properties during a cold period in February 2012 at the Puy de Dôme station, as well as LIDAR measurements (at the Cezeaux site) in France. A change in aerosol physical and chemical properties was observed in the free troposphere with an increase in aerosol mass concentrations, number of larger particles, and presence of organic and nitrate particles.

Aerosol size distributions over a 20-month period at Mt. Tai station in central east China were presented by Shen *et al.* (2016). Data was separated by planetary boundary layer influence or free tropospheric by time of day. When new particle event days were excluded, the aerosol number concentration showed a seasonal variation with maximum

in summer and minimum in winter, which was influenced by seasonal variations in the planetary boundary layer evolution and by air mass advection.

Sharma *et al.* (2016) investigated the unique boundary layer at Mauna Loa Observatory (MLO) with a combination of radiosondes launched from the observatory and a novel aerosol profiling technique called CLidar or camera lidar. The authors found that that high altitude resolution and near-ground capabilities of the CLidar technique were particularly useful to study the complex MLO boundary layer, which is influenced by a combination of off-island (free tropospheric) winds and radiation winds (from heating and cooling).

Aerosol and Trace Gas Climatology

Eleven papers within this special issue utilized aerosol and/or trace gas properties observed at a high elevation to gain insight into trends, temporal patterns, processes and source regions of atmospheric constituents. All papers included within this theme present results based on long-term (i.e., multi-year) data from mountain sites.

Bukowiecki *et al.* (2016) summarized aerosol properties from the Jungfraujoch in Switzerland using more than 20 years of measurements obtained from numerous in-situ studies. This climatology includes physical, chemical and optical aerosol properties as well as aerosol-cloud interactions and cloud characteristics. Both aerosol size distribution and the aerosol chemical composition were shown to be fairly constant in time due to the long distance from aerosol sources, and, because of this consistency, climate relevant aerosol properties can be derived from these properties. Thirteen years of measurements of ultrafine (3–10 nm diameter) aerosols from Storm Peak Laboratory in Colorado were presented in Hallar *et al.* (2016). Previous work has shown that frequent new particle formation occurs regularly at the site, and this long-term climatology of ultrafine aerosols clearly demonstrates a seasonal dependence on new particle formation at Storm Peak Laboratory, with a maximum during the spring season and a minimum in summer. Recent sulfur dioxide data indicates a strong source region west of Storm Peak Laboratory, and corresponding to the predominant wind direction observed during new particle formation events.

Schwab *et al.* (2016a, b) and Brandt *et al.* (2016) are three companion papers describing measurements at Whiteface Mountain in the Andirondack Mountain region in northern New York. Schwab *et al.* (2016a) presents a historical overview of the site and its measurements, making the case for the utility of long-term monitoring while the other two papers provide more detailed findings about the aerosol, cloud, and precipitation measurements (Schwab *et al.*, 2016b) and ozone and trace gases (Brandt *et al.*, 2016). In particular, both the aerosol and gas phase measurements collected over a period of 40 years at Whiteface Mountain highlight the past success of air pollution regulation under the U.S. Clean Air Act and Amendments and pave the way for future progress in reducing air pollution.

Sheridan *et al.* (2016) updated the long-term wind and aerosol climatology for the high elevation Atmospheric Research Observatory at the Amundsen-Scott South Pole Station, due to evaluate whether recent increased human activity at the station was observable in the long-term time series. Results show that the long-term wind speed and direction measurements have not changed appreciably, and no significant trends were observed in particle number concentration, aerosol light scattering coefficient, or any aerosol parameter except scattering Ångström exponent, over the 36-year record.

Kirchner *et al.* (2016) examine persistent organic pollutants at several high elevation sites in Europe and find broadly similar patterns and sources for three different high elevation sites. Importantly they find a downward trend in concentrations of the pesticide endosulfan. Chambers *et al.* (2016) describe a new, simplified method to identify baseline air masses, compared to more regionally influenced air masses using high sensitivity measurements of Radon. They apply this method to data from three GAW stations; Jungfraujoch, Mauna Loa and Cape Grim and demonstrate the suitability of the method by comparing baseline CO_2 mixing ratios using this method. Leuchner *et al.* (2016) report on formaldehyde observations made at the high elevation Zugspitze site in Germany. They report the first background, long-term observations with an emphasis on the sources and seasonal maximum, observed in summer.

Zhu *et al.* (2016) presented a climatology (both seasonal and diurnal) of equivalent black carbon (EBC) at a site at high altitude site on China's Tibetan plateau. Using back-trajectory analysis they were able to link peaks in observed concentration with middle to long range transport from more polluted regions. A similar approach was used by Kang *et al.* (2016) to identify source regions influencing the observed seasonality of various elements and their enrichment factors for a different location on the Tibetan plateau.

Model Evaluation

Due to the inherent complex terrain of mountain sites, these locations offer an opportunity and challenge for model comparisons. Two papers within this special issue focused on model and measurement intercomparison at mountain sites.

Model simulated number concentrations of condensation nuclei larger than 10 nm in 2009 were compared with those measured at the Storm Peak Laboratory, in northwestern Colorado, by Yu *et al.* (2016). Vertical profiles and seasonal variations of key parameters controlling particle formation and growth were investigated. The model overall captures absolute values aerosol concentration and their variations, especially in the spring months where regional scale new particle formation occurred frequently.

Barbet *et al.* (2016) utilized the WRF-Chem model at high resolution (10 km spacing) to interpret aerosol composition data, from a compact Time-of-Flight Aerosol Mass Spectrometer, at the Puy de Dôme research station in France They found that the high-resolution model was able to capture the diurnal variability in organic aerosol measured by in-situ instrumentation although the model underestimated organic concentrations due to issues with the parameterization of secondary organic aerosol formation.

CONCLUDING REMARKS

The 2nd Atmospheric Chemistry and Physics at Mountain Sites Symposium was a successful international workshop that attracted thirty-six participants who exchanged the latest scientific information and enhanced their knowledge about various aspects of high elevation atmospheric measurements. The special issue publishes 34 papers selected from the numerous conference presentations. This overview provides some highlights of the scientific findings described in these papers. The guest editors would like to thank the authors for their contributions and the referees for their thorough reviews and critical comments. To continue the successful collaborations established by the 1st and 2nd workshops, the researchers are looking forward to the 3rd Atmospheric Chemistry and Physics at Mountain Sites Symposium that will be held in Japan in 2017.

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