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On the Limits to Manage Air-Quality in Glacier Bay

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Abstract

In Glacier Bay National Park, about 95% of the visitors come on board of cruise ships. The National Park Service has the mandate to manage park resources like air quality and visibility, while ensuring visitation. To understand the impact of cruise-ship emissions on the overall concentrations in Glacier Bay, emission-source contribution ratios (ESCR) and the interaction of pollutant from local and/or distant sources were determined using results from four WRF/Chem simulations of the 2008 tourist season (May 15 to September 15). These simulations only differed by the emissions considered: Biogenic emissions only (CLN), biogenic plus activity-based cruise-ship emissions (REF), biogenic plus all anthropogenic emissions except cruise-ship emissions (RETRO), and all aforementioned emissions (ALL). In general, ESCRs differed among pollutants. Interaction between pollutants from cruise-ship emissions and species from other sources including those advected into the bay decreased towards the top of the atmospheric boundary layer. Pollutants from different sources interacted strongest (lowest) in the west arm of the fjord where ships berthed for glacier viewing (in areas of the bay without cruise-ship travel). Pollutant interaction both enhanced/reduced NO_2 concentrations by 10% (4 - 8 ppt absolute). Except for ozone, cruise-ship emissions on average governed air quality in the bay. On days with cruise-ship visits, they contributed between 60% and 80% of the bay-wide daily mean SO_2 and NO_2 concentrations below 1 km height. On days without visits, cruise-ship contributions still reached 40% due to previous visits. Highest cruise-ship ESCRs occurred during stagnant weather conditions. Despite the fact that all coarse particulate matter was due to anthropogenic sources, worst visibility conditions were due to meteorology. The results suggest limits as well as windows for managing air quality and visibility in Glacier Bay.

Keywords

Glacier Bay National Park, Emission-Source Contribution, Cruise-Ship Emissions, Air-Quality Management, Interaction of Pollutants

1. Introduction

In recent years, the phenomenon of "last chance tourism" has increased. Herein people wish to visit places such as the Arctic, Antarctic, and tidewater glaciers, which they anticipate to be irreversibly impacted by climate change, before they are gone [1] [2] [3]. Cruise-ship tourism capitalizes on this desire, and targets areas with accessible glaciers. During their voyage, however, cruise ships emit primary particles and precursor gases such as sulfur dioxide (SO_2), nitrogen oxides (NO_x = NO + NO_2 nitric oxide and nitrogen dioxide), and volatile organic compounds (VOC) [4] [5] in the marine and coastal atmospheric boundary layer (ABL). Consequently, large cruise ships may be the major or, at times the only anthropogenic emission source in these remote areas.

Once these primary pollutants and particles are in the ABL, they form secondary pollutants and/or secondary particles by chemical reactions and gas-to-particle conversion [6]. In high latitudes and fjords, pollutants and particles often accumulate in the ABL when they become trapped during the frequent, sometimes multi-day inversions [7] [8] [9]. On relatively warm days, high NO_2 concentrations may become visible as a brown layer underneath the inversion or at the top of the ABL. When inversions coincide with high relative humidity, particles swell and become visible as haze [10] [11] resulting in reduced visibility [12]. Thus, pollutants emitted by cruise ships can negatively impact tourists' experience, visibility and park resources.

Glacier Bay National Park is located in southeastern Alaska, and represents a coveted destination for cruise-ship passengers. The National Park Service (NPS), which manages Glacier Bay by regulating vessel volume and operating conditions, has a dual mandate to both promote visitation while also protecting park resources and values. Glacier Bay has a number of accessible tidewater glaciers, but no roads that allow visitors to experience and enjoy these and other park resources. Thus, cruise ships play a crucial role in providing visitor access, regularly constituting over 95% of the >450,000 annual visitors. The NPS must thus carefully consider the value of cruise ships for meeting the visitation mandate with the impacts from cruise ships that may violate the resource-protection mandate, particularly to visibility, air quality and other park resources.

Unfortunately, the atmosphere knows no boundaries. Hence, unlike evaluating water-quality impacts, where the inputs of a pollutant can be calculated relative to the volume of the receiving water body to compare with national concentration standards, the atmosphere prohibits assessing impacts in terms of contaminant emissions into a closed volume. Instead air quality is compared to the National Ambient Air Quality Standard (NAAQS), which is expressed as a mean concentration per volume (usually 1 m^{-3}) or as a fraction of a particle number (e.g. parts per million (ppm), part per billion (ppb)) threshold for a determined amount of time that varies by pollutants.

Even when an inversion extends over the entire bay, and limits the exchange with air from aloft, still some lateral exchange may occur at the park's entrance to Icy Strait [9], which is heavily used by large ships. Emissions from sources in this area can thus affect air quality in the bay.

Under inversion conditions, inversion height varies over the bay [9] and/or inver-

sions may be limited to certain areas. Advection of contaminants emitted elsewhere can thus influence the contaminant concentrations in Glacier Bay in addition to the contaminants emitted during cruise-ship visits in Glacier Bay [9]. On the other hand, pollutants from cruise-ship emissions in the bay may be transported out of the bay as well.

Knowledge on how pollutants from local and distant sources interact to affect air quality is limited. During the Arctic Research of the Composition of the Troposphere from Aircraft and Satellites (ARCTAS), and Aerosol, Radiation, and Cloud Processes affecting Arctic Climate (ARCPAC) campaigns in April, June, and July 2008, carbon monoxide (CO) concentrations from anthropogenic emissions in Asia and Europe were found in the mid-troposphere of the North American Arctic [13]. The North American Arctic mid-troposphere received black carbon (BC) from biomass burning in Russia, while polluted air from Asian anthropogenic sources occurred in the upper troposphere [14].

Similar results have been found with marine-sourced emissions affecting air quality over terrestrial areas. For example, air-quality model simulations revealed that in northern Germany and Denmark, more than 50% of the summer 2000 sulfate, nitrate and ammonium aerosol concentrations were due to ship emissions in the North Sea [15]. The Sulfur Emission Control Area implemented in 2009 in the North Sea reduced SO_2 and sulfate-aerosol concentrations, but slightly increased nitrate-aerosol concentrations [15]. In summer 2012, as part of the European Arctic Climate Change, Economy, and Society (ACCESS) project, a field campaign took place off the coast of Norway. A major goal was to quantify the contributions of emissions from regional shipping and offshore gas and oil production as well as distant emission sources (e.g. smelting on the Kola Peninsula, Siberian biomass burning) on local air pollution in the Norwegian Arctic [16]. Air-quality model simulations demonstrated that along the Norwegian coast, local ship emissions increased the 15-day mean near-surface concentrations of NO_x, ozone (O_3), BC and particulate matter of 2.5 μm or less in diameter ($PM_{2.5}$) up to 80%, 5%, 40% and 10%, respectively [17].

Given these results, and the dual mandate of the NPS, we examined the limits to which the NPS can manage/control air quality successfully in Glacier Bay. While the NPS can regulate the number of cruise ships that enter the park, set entrance quota and/or speed limits, and set up competitive contracts that result in ships using low sulfur fuel [18], the NPS cannot control inversions, precipitation, biogenic emissions, or the advection of pollutants from emissions outside of the park boundaries.

We thus investigated how pollutants from different local and distant sources contribute to air quality and their overall impact on visibility in Glacier Bay and how these pollutants interact. The goal was to examine whether and to which degree air quality and visibility in Glacier Bay are determined by emissions within the bay. To achieve this goal, we 1) identified impacts from distant sources on concentrations in Glacier Bay over the length of a tourist season (May 15 to September 15); 2) quantified the contributions from different sources (natural emissions and background, cruise ship emissions, and anthropogenic emissions except cruise-ship emissions, all of these) on

air quality and visibility conditions in the bay; 3) examined whether pollutants from different local and distant sources interacted with each other, thereby increasing/reducing concentrations of products/reactants, and 4) compared the contributions of emission sources to the mean concentrations over Southeast Alaska and Glacier Bay. An important pre-requisite for any air-quality management is that air-quality is governed by local controllable sources. Thus, we used a well-evaluated air-quality model and performed four simulations that only differed by the emissions considered. The setup of the simulations was designed to determine the emission-source contribution ratios (ESCR) [19] of the various sources and the interaction [20] [21] between pollutants from various sources in Glacier Bay and Southeast Alaska. In addition, we compared the Southeast Alaska and bay-wide mean concentrations in the ABL as they should differ when cruise-ship emissions are the main source for pollutants in the bay.

2. Experimental Design

2.1. Model Description

The air-quality model used in our study is the WRF/Chem [22] [23]. It was set up identical to [11]: Cloud processes on the resolvable and subgrid-scale were calculated by the WRF-Single-Moment 5-class cloud-microphysics scheme [24] and a further-developed version of [25]'s ensemble convective scheme, respectively. The Goddard two-stream multi-band scheme [26] and Rapid Radiative Transfer Model [27] served to determine shortwave and long-wave radiation including cloud and aerosol-radiation feedbacks [28]. Surface and atmospheric boundary layer physics were treated in accord with [29]. A modified version of the so-called NOAH land-surface model [30] calculated the exchange of heat and matter at the surface-atmosphere interface, as well as the snow, soil-temperature and soil-moisture and frozen ground conditions. Gas chemical processes were calculated by the Regional Acid Deposition Model version 2 chemical mechanism [31] and used inline calculated photolysis rates [32]. Aerosol dynamics, physics, and chemistry were considered by the Modal Aerosol Dynamics Model for Europe [33] and Secondary Organic Aerosol Model [34].

2.2. Initialization and Boundary Conditions

All simulations were driven by the 1˚ × 1˚, 6 h-resolution National Centers for Environmental Prediction global final analyses (FNL) data [35] as lateral boundary conditions for the meteorological fields. The FNL snow, soil-temperature and moisture conditions, as well as sea-surface temperatures (SST) served to initialize the meteorological and surface fields. All simulations ran in "forecast mode" with re-initialization of the meteorology every five days. Note that from a modeling standpoint, the simulations could have been run using reanalysis data instead of analysis data, and in nudging mode rather than forecast mode. We chose the forecast mode because managers have experience with the uncertainty associated with weather forecasts. Weather forecasts base on numerical weather prediction (NWP) that uses analysis data for initialization to produce a forecast. Thus, performing the study in forecast mode provides a framework

of "known performance" for stakeholders.

In all four WRF/Chem simulations, the same idealized profiles of clean air background concentrations served as initial conditions of the chemical fields at the start of the simulation and provided the lateral boundary conditions over the May 15 to September 15, 2008 "tourist season" [11].

The model domain encompassed the atmosphere over Southeast Alaska with 28 layers from the surface to 100 hPa, and 120 × 120 grid-points of 7 km horizontal increment centered at 58.5N, 135.5W. To permit adjustment of the meteorological and chemical fields and minimize errors from lateral boundary effects, we discarded five grid-points on each lateral boundary from the results (**Figure 1**).

2.3. Emission Data

Anthropogenic emissions, except from cruise ships were derived from the 0.5˚ × 0.5˚ Reanalysis of the Tropospheric Chemical Composition (RETRO) data. For cruise ships, we calculated activity-based emissions using Automated Information System (AIS) voyage data (ship position, cruise speed, operation mode) and the individual ships' characteristics (engine power, size, fuel type, maximum cruise speed, etc.) [11]. We refer to this emissions inventory as REF. The emissions inventory ALL combined the RETRO and REF inventories (**Figure 1**). WRF/Chem calculated the biogenic emissions (e.g. NO_x, various VOCs) inline depending on vegetation and soil type, soil temperature and moisture, and atmospheric conditions following [36] [37] in all simulations.

2.4. Simulations

We performed four WRF/Chem simulations for the tourist "season". For all simulations, model setup, boundary conditions, and initialization were identical except for the choice of the emissions inventory.

Our CLN simulation included only biogenic emissions and, due to the absence of anthropogenic emissions, represented a "clean" atmosphere [38]. Our reference (REF) simulation included the biogenic emissions plus the activity-based cruise-ship emissions. Evaluation of this simulation with respect to various aspects can be found in [9] [11]. The RETRO simulation included biogenic emissions and other anthropogenic emissions including commercial shipping, but no cruise-ship emissions. Finally, the ALL simulation represented the atmospheric composition of the 2008 tourist season as it included activity-based cruise ship and other anthropogenic emissions as well as biogenic emissions.

2.5. Analysis

Being in control of the air quality in Glacier Bay would require that in the bay, air quality only or at least to a high percentage depends on the emissions occurring inside the bay. This means that advection of pollutants from other sources including cruise ships outside of Glacier Bay would have to be negligibly small relative to the pollutant concentrations from sources occurring inside the bay. Furthermore, the concentrations inside

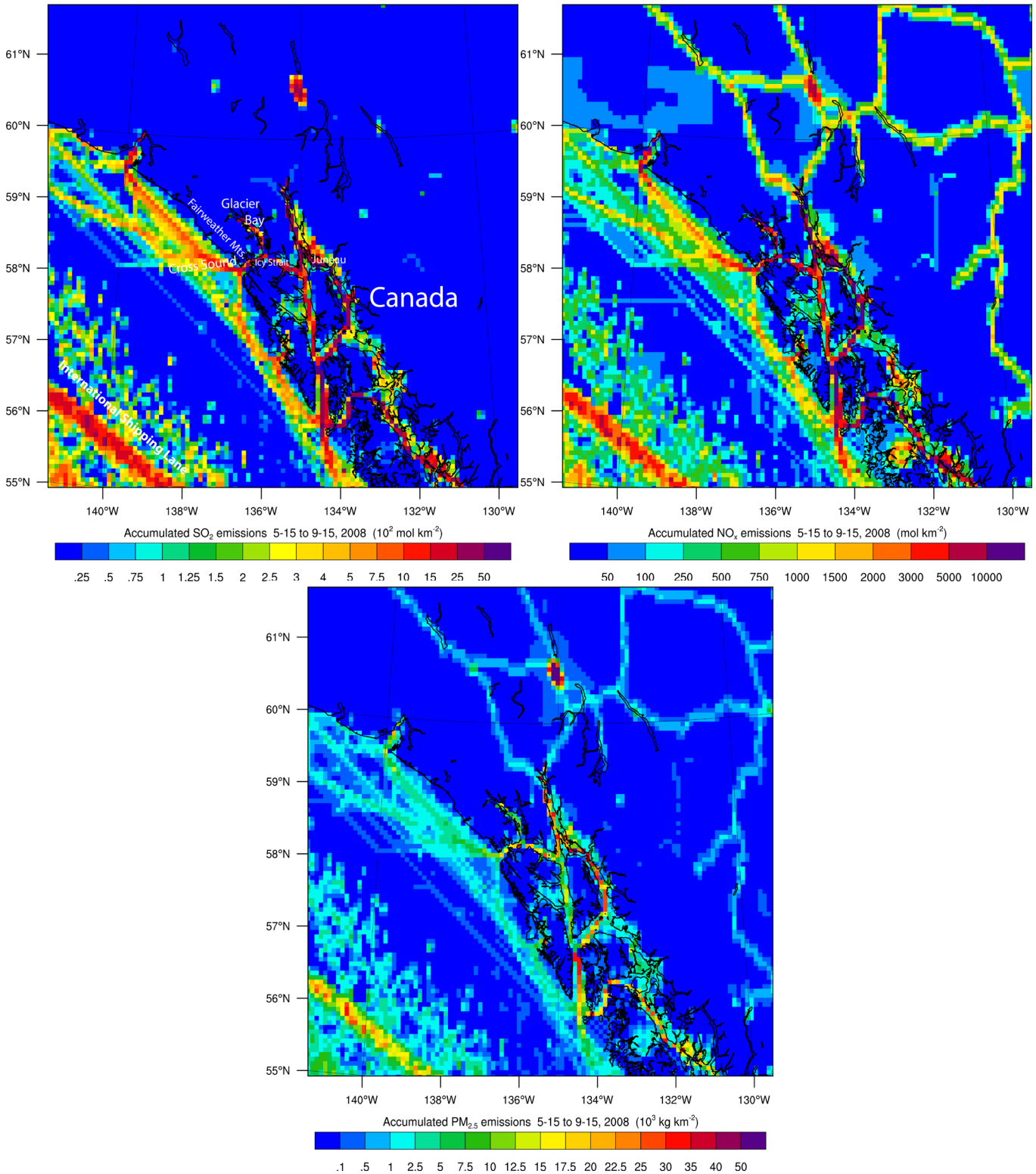


Figure 1. Season accumulated anthropogenic emissions of (upper left to bottom) SO_2, NO_x, and $PM_{2.5}$ in southeast Alaska as considered in ALL. Legends differ among panels. For season accumulated emissions of REF (cruise ships only) see **Figure 1** in [11]. Emissions along the coast and on the waterways are mainly due to cruise ships, while those in the southwestern corner of the domain are due to commercial shipping other than cruises. No anthropogenic emissions occur in CLN.

the bay should not have been modified by chemical reactions with pollutants from local non-controllable and/or distant sources.

To test the hypothesis of *No interaction of pollutants from different local and/or distant sources* we applied the principle of superposition [20] [21]. Applied to our task it means that the sum of the concentrations of the simulations considering only one type of emission sources should equal the concentrations of the simulation that considered all emission sources (ALL).

Since both REF and RETRO considered background concentrations and biogenic emissions, we subtracted the concentrations obtained by the CLN simulation from both REF and RETRO to account for the contribution of natural emissions and background concentrations to the total atmospheric concentrations only once. After some algebra, we obtain $C_{i,j,k,t,m}(\mathrm{REF}) + C_{i,j,k,t,m}(\mathrm{RETRO}) - C_{i,j,k,t,m}(\mathrm{CLN})$, which we compared with the concentration of $C_{i,j,k,t,m}$(ALL)

$$C_{i,j,k,t,m}(\mathrm{REF}) + C_{i,j,k,t,m}(\mathrm{RETRO}) - C_{i,j,k,t,m}(\mathrm{CLN}) = C_{i,j,k,t,m}(\mathrm{ALL}) \quad (1)$$

Here, i, j, k are the indices of a model grid-cell, m is the chemical species or particulate matter, and t is time.

If there were no interactions of pollutants from different local and/or distant emission sources, the left hand side of Equation (1) would not differ significantly, at the 95% confidence level in a two-tailed t-test [39] from the concentration $C_{i,j,k,t,m}$(ALL) on the right hand side. Any difference between the left and right hand side of Equation (1) can be expressed as a fraction, α, stemming from interactions of pollutants from the three groups of sources (biogenic emissions and background, cruise-ship emissions, anthropogenic emissions other than cruise-ship emissions).

Due to atmospheric transport, the atmospheric composition can contain pollutants emitted at different places. For example, assume a wind of 10 $m \cdot s^{-1}$ blows from Juneau to Glacier Bay that are about 150 km apart. Pollutants emitted in Juneau by anthropogenic sources including cruise ships would reach Glacier Bay about 4.2 hours later. During transport, chemical reactions and gas-to-particle conversion of the primary pollutants and particles emitted at Juneau produce secondary pollutants and particles. Furthermore, chemical reactions, removal processes, and emissions from sources along the way, as well as mixing processes modify the air composition. Once the aged air reaches Glacier Bay, reactions between these pollutants from various different distant sources and those from the local cruise-ship emissions may occur. Following [20] [21] the fraction $\alpha_{i,j,k,m,t}$ to the concentration of ALL due to interaction of species from different emission sources can be determined as

$$\frac{C_{i,j,k,t,m}(\mathrm{ALL})}{C_{i,j,k,t,m}(\mathrm{ALL})} - \frac{C_{i,j,k,t,m}(\mathrm{REF})}{C_{i,j,k,t,m}(\mathrm{ALL})} - \frac{C_{i,j,k,t,m}(\mathrm{RETRO})}{C_{i,j,k,t,m}(\mathrm{ALL})} + \frac{C_{i,j,k,t,m}(\mathrm{CLN})}{C_{i,j,k,t,m}(\mathrm{ALL})} = \alpha_{i,j,k,t,m} \begin{cases} > 0 & \text{enhancement} \\ \cong 0 & \text{no interaction} \\ < 0 & \text{diminuation} \end{cases} \quad (2)$$

The emission-source contribution ratios (ESCR) [19] of the various emission sources,

i.e. natural/biogenic, cruise ships, the collective anthropogenic emissions except cruise ships, and all anthropogenic emissions to the ALL concentrations reads

$$\begin{aligned} 100\% &= 100 \cdot \left(\frac{C_{i,j,k,t,m}(ALL)}{C_{i,j,k,t,m}(ALL)} \right) \\ &= \left[\left(\frac{C_{i,j,k,t,m}(REF) - C_{i,j,k,t,m}(CLN)}{C_{i,j,k,t,m}(ALL)} \right) + \left(\frac{C_{i,j,k,t,m}(RETRO) - C_{i,j,k,t,m}(CLN)}{C_{i,j,k,t,m}(ALL)} \right) \right. \\ &\quad \left. + \frac{C_{i,j,k,t,m}(CLN)}{C_{i,j,k,t,m}(ALL)} + \alpha_{i,j,k,t,m} \right] \cdot 100\% \\ &= \left(ESCR_{CLN} + ESCR_{REF} + ESCR_{RETRO} + \alpha \right) \cdot 100\% \end{aligned} \quad (3)$$

Here $C_{i,j,k,t,m}(CLN)/C_{i,j,k,t,m}(ALL) = ESCR_{CLN}$ is the fractional contribution from natural/biogenic emissions and the background concentrations to the overall concentration $C_{i,j,k,t,m}(ALL)$ for i, j, k, t, and m. This contribution refers to the natural background chemistry for which it is always positive.

Furthermore, $\left(C_{i,j,k,t,m}(REF) - C_{i,j,k,t,m}(CLN)\right)/C_{i,j,k,t,m}(ALL) = ESCR_{REF}$ is the emission source contribution ratio to the ALL concentrations due to cruise ships. The emission source contribution ratio $\left(ESCR_{RETRO} = \left(C_{i,j,k,t,m}(RETRO) - C_{i,j,k,t,m}(CLN)\right)/C_{i,j,k,t,m}(ALL)\right)$ represents the non-cruise ship related anthropogenic emissions. Multiplication of α and the $ESCR_x$ (x = CLN, REF, RETRO) terms by 100% provides the contribution of interaction and the respective emission sources to the concentration in ALL, $C_{i,j,k,t,m}(ALL)$ in percent.

According to Equation (2), interaction of pollutants from different local and distant sources, and non-linear chemical reactions can enhance or diminish ALL concentrations as compared to the concentrations obtained by adding the concentrations in response to the individual emissions inventories.

Equation (3) permits negative ESCR values, when the concentration in CLN exceeds that of REF or RETRO, respectively. Negative $ESCR_x$ (x = REF, RETRO) values result from interaction. They mean that the emissions from the respective source permitted chemical reactions and processes that reduced the ALL concentration of the respective species. In other words, if the emitted species would not have reacted, the ALL concentration would be higher than it is by the absolute of the percentage. Adding reactive species m to a (clean) air sample reduces the species it reacts with and increases the concentrations of the reaction products. The products may further react when they are not an end product of the reaction chain. When the species m and its initial reactant(s) are not reproduced in the reaction chain, their concentrations decrease [6].

In general, reactions requiring low energy are favored over those needing comparatively higher energy at same temperature. Reaction rates differ among reactive species. Furthermore, reactions may be limited by low concentrations of their reactants. For instance, in a closed air parcel of NO-NO_2-O_3 under quasi-steady state conditions, the ozone concentration remains constant despite of the occurring reactions. Adding NO_x to this air parcel decreases the O_3 concentration. Adding VOC instead introduces competing reactions. As long as the VOC/NO_x ratio of the parcel is low, its air is VOC-sen-

sitive. It is NO_x sensitive for low NO_x and high VOC concentrations. In this case, O_3 concentration increases with increasing NO_x and little response of VOC.

In the clean atmosphere, background chemistry also occurs [6]. Vegetation emits various types of VOCs [36] [40] that may differ strongly in their reaction rates [41]. Emission rates depend on plant-available water in the root zone, air temperature and moisture, and photosynthetic active radiation, among other things [37] [42]. Soil bacteria emit NO_x as part of their metabolism that depends on soil temperature and moisture [36] [37] [40] [42]. Note that WRF/Chem considers these processes [22] [23].

Chemical processes conserve the total mass of atoms, but not molecule species. Anthropogenic emissions may add species that also have natural sources and/or species that do not occur in the clean atmosphere [6]. Consequently, the competition for reactants changes and interaction of species from natural and anthropogenic sources and/or different anthropogenic sources may occur. In the case of cruise-ship emissions, for instance, both faster and slower reacting VOCs join the natural VOCs. As illustrated in the above example of the reaction system NO_x-O_3-VOC, anthropogenic emissions may shift the chemical regime leading to reduced concentrations even of emitted species. The chemical regime also alters the concentrations of secondary pollutants, *i.e.* those pollutants that form by reaction of the primary, *i.e.* emitted pollutants. Finally, and with respect to managing park resources, it is worth mentioning that anthropogenic emissions also change the fractional composition of particles. However, particles of different composition have different impacts on visibility [12].

We examined the mean ESCRs from the different sources and pollutant interactions to the atmospheric composition of ALL to assess the degree to which NPS managers have the possibility to manage air quality in Glacier Bay. We used comparison to the mean concentrations in Southeast Alaska with the assumption that differences between the mean composition of ALL in Southeast Alaska vs. Glacier Bay provide evidence that local emissions within the bay's air shed govern its air quality. The value of $ESCR_{REF}$ represents the degree to which cruise-ship emissions in Glacier Bay determine the concentration of the respective species inside the bay. The difference 1-$ESCR_{REF}$ is the limit for management. The lower this limit is for an emitted species (1-$ESCR_{REF}$ $\ll$ $ESCR_{REF}$), the greater are the possibilities of the NPS for managing the pollutant by emission-control measures.

3. Results

3.1. Primary Pollutants

Anthropogenic emissions contributed all coarse particulate matter, *i.e.* PM_{10} with diameters exceeding 2.5 μm, but being less than or equal to 10 μm in diameter in ALL, RETRO, and REF. In CLN, all PM_{10} was $PM_{2.5}$. Comparison of the REF and RETRO seasonal mean profiles of SO_2 over the Glacier Bay water proper (**Figure 2**) reveals that transport of pollutants from other sources than cruise ships contributed to the ALL concentrations in the maritime ABL, and up to 700 hPa (~3 km above sea level). The SO_2 peak around 1 km (900 hPa) was related to frequent inversions. The second peak at

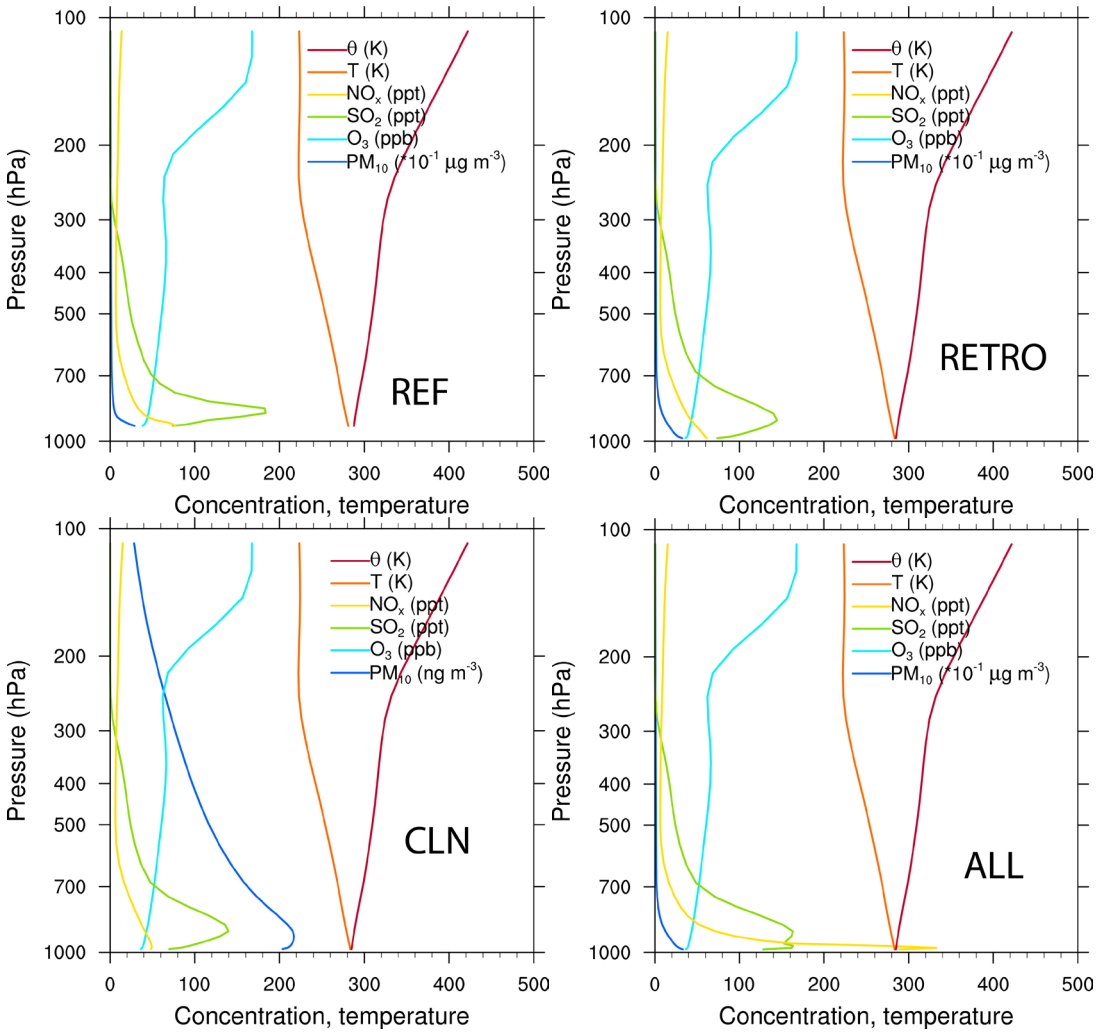


Figure 2. Glacier Bay bay-wide season mean vertical profiles for selected trace gas, and particulate matter concentrations, air and potential temperature as obtained by the REF, RETRO, CLN, and ALL simulations. The unit for PM_{10} in CLN differs from that in the other panels. In CLN, all PM_{10} was $PM_{2.5}$.

about 1.5 km above water level (850 hPa) relates to the background profile (compare to CLN).

The height of highest daily mean SO_2 concentrations in the ABL differed with time due to local weather conditions (not shown; cf. [9]). On days with low inversion heights, highest concentrations in Glacier Bay occurred below 1 km. When bay-wide, highest daily mean SO_2 concentrations existed at about 850 hPa at the top of the ABL, SO_2 concentrations were low close to the water. This finding points to large-scale forcing with vertical mixing and/or convection on those days.

Comparison of bay-wide seasonal means of CLN, REF, RETRO, and ALL NO_x concentrations (**Figure 2**) supports the above finding that pollutants from emissions outside the bay affected the ALL concentrations below 700 hPa. Seasonal bay-wide mean O_3 profiles barely differed indicating that they mainly represent O_3 background concentrations.

Averaged over the season and to about 152 m above the water surface, ALL SO_2 emissions and concentrations correlated significantly at the 95% confidence level over the international shipping lane (southwestern corner of the model domain), port cities, some waterways, and most of Canada (Figure 3). In areas without anthropogenic emissions, for instance the glacier-covered mountains, absence of emissions correlated with

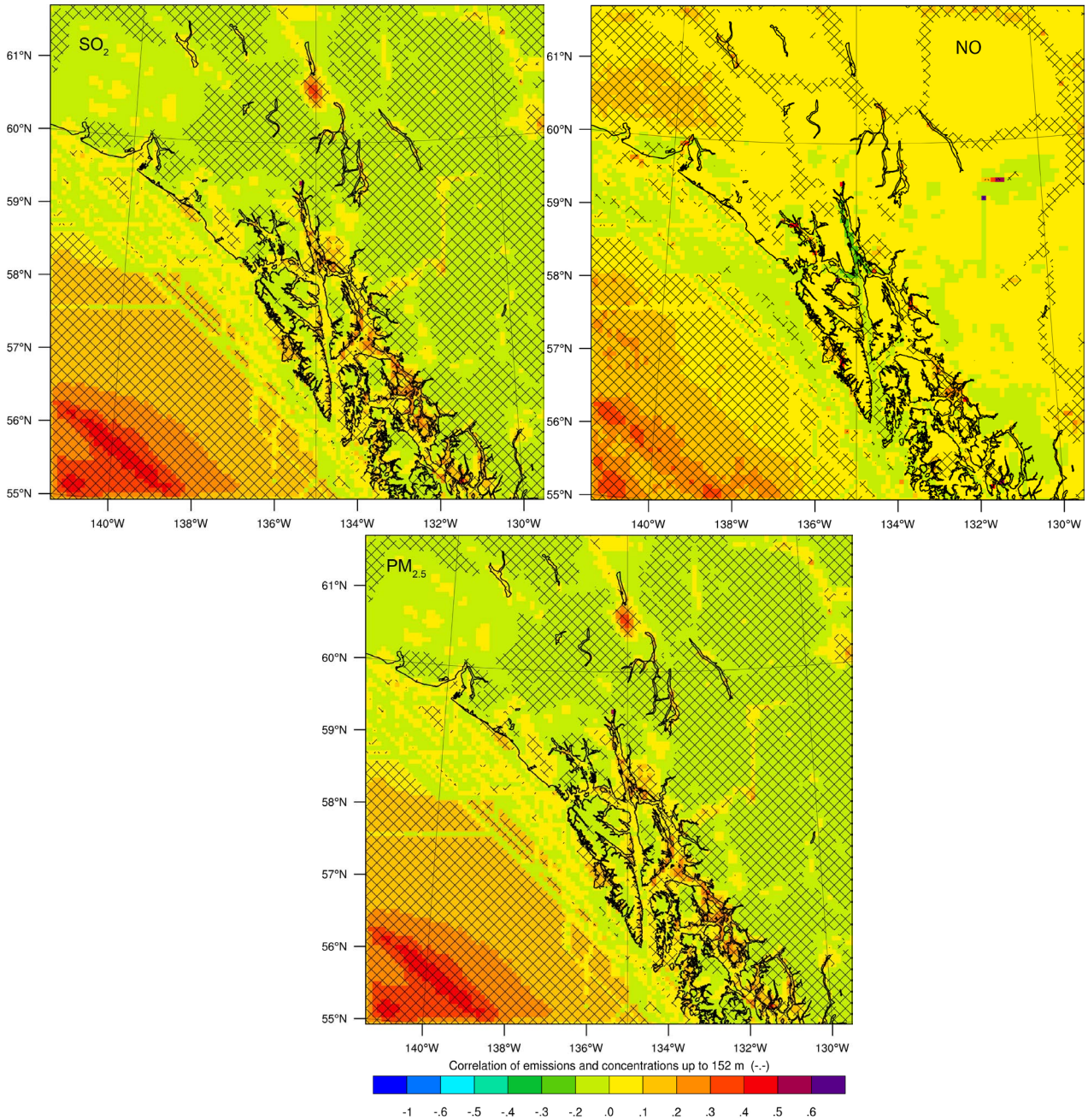


Figure 3. Correlations between ALL emissions and ALL concentrations in the first 152 m above the surface over the tourist season for SO_2, NO, PM_{10}, and $PM_{2.5}$. Shaded areas indicate significant correlation at the 95% confidence level. Note that this plots also includes correlations of no emissions with the concentrations in the first 152 m or so. In this case, a significant correlation means that the area is only marginally affected by advection of pollutants from other regions.

low concentrations. Below the first 152 m or so, significant, but low NO emission-concentration correlations occurred over the major highways in Canada. NO emission-concentration correlations were comparatively higher over the waterways in the Alexander Archipelago including Glacier Bay, locally along the coast where cruise-ships travel density was high (cf. [43]), and reached correlation coefficients about 0.5 over the international shipping lane (compare **Figure 1**, **Figure 3**).

Across Southeast Alaska waters, ALL PM_{10} concentrations visibly reflected the ship routes as ships are the only source for coarse particulate matter over water (**Figure 4**). High SO_2 concentrations occurred in Icy Strait and just up the west side near the entrance of Glacier Bay park (**Figure 3**). Here cruise-ship speeds were higher than inside Glacier Bay. Note that under certain synoptic conditions, pollution was transported from Icy Strait into Glacier Bay elevating pollutant concentrations [9].

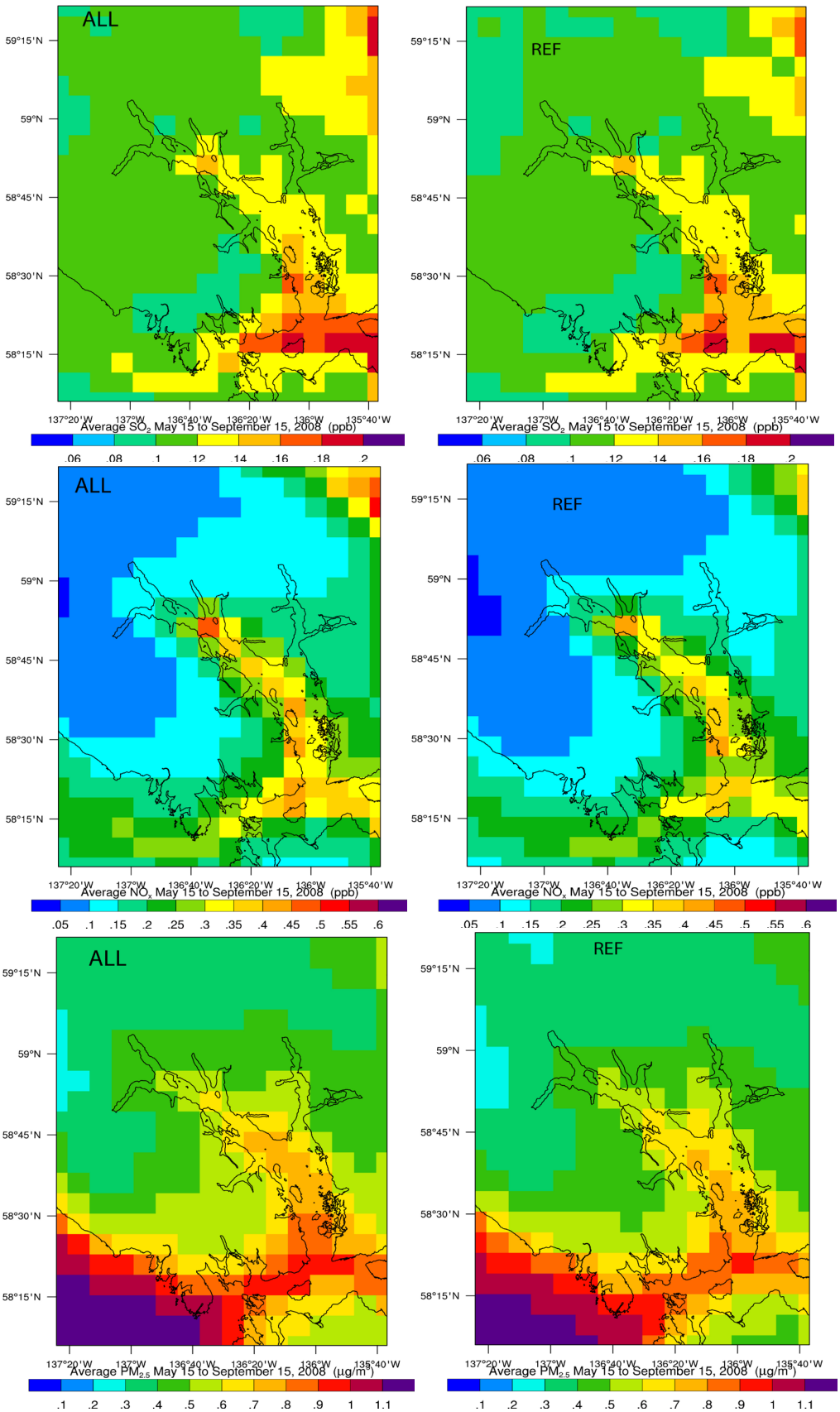
In ALL and REF, the cruise path thru the bay was clearly visible at the time of the voyage for all emitted species, and even notable on season average for NO_x and $PM_{2.5}$ (**Figure 4**). In Glacier Bay, seasonal mean REF and ALL SO_2, and NO_x concentrations were highest in the lower inlet downstream of where cruise ships berth for several hours for glacier viewing, while running their auxiliary engines on low load to generate energy for hoteling [11]. Note that at low loads and during maneuvering, combustion is often incomplete meaning high emissions [4] [44] [45].

Highest concentrations did not exactly correspond to the areas adjacent to the tidewater glaciers where ships berth for several hours to allow passengers time to experience the scenery. The slightly off-set of the "expected location" for concentration maxima was due to the coarse grid resolution, discretization of the advection-diffusion equations, and lost AIS signals. The former two are well-known modeling artifacts [41] [46] [47].

Comparison of CLN and RETRO SO_2 concentrations revealed marginal increases in Glacier Bay due to advection of SO_2 from other anthropogenic sources than cruise ships (not shown). Comparing SO_2 concentrations simulated by REF and ALL confirmed these findings (**Figure 4**). The same was true for the NO_x concentrations in Glacier Bay. We conclude that cruise-ship emissions governed the magnitude and distribution of SO_2 and NO_x concentrations in the bay most of the time.

Over the bay, increases in ALL and REF hourly NO_x and hence daily mean concentrations corresponded to cruise-ship visits (**Figure 5**). The highest NO_x concentrations occurred when two large cruise ships visited the bay on the same day during strong inversions [9]. Since peroxide-acetyl-nitrate (PAN) is a reservoir for NO_x, NO_x concentrations increased at the cost of PAN when temperature increased, and *vice versa* [38].

VOC has both anthropogenic and natural sources [6] [36] [37] [41]. In all four simulations, biogenic emissions governed the VOC concentrations over land in Southeast Alaska. In Glacier Bay National Park, the total amount of biogenic emissions is low because glaciers and water cover large areas of the park. Comparison of RETRO and CLN VOC concentrations showed marginal increases in RETRO VOC over Glacier Bay. This finding indicates that advection of anthropogenic VOC only marginally modified the


ALL
REF
59°15'N
59°N
58°45'N
58°30'N
58°15'N
137°20'W
137°W
136°40'W
136°20'W
136°W
135°40'W
Average SO2 May 15 to September 15, 2008 (ppb)
.06 .08 1 12 14 .16 .18 2
Average NO, May 15 to September 15, 2008 (ppb)
.05 .1 .15 .2 .25 .3 .35 .4 .45 .5 .55 .6
Average PM2.5 May 15 to September 15, 2008 (µg/m3)
.1 .2 .3 .4 .5 .6 .7 .8 .9 1 1.1

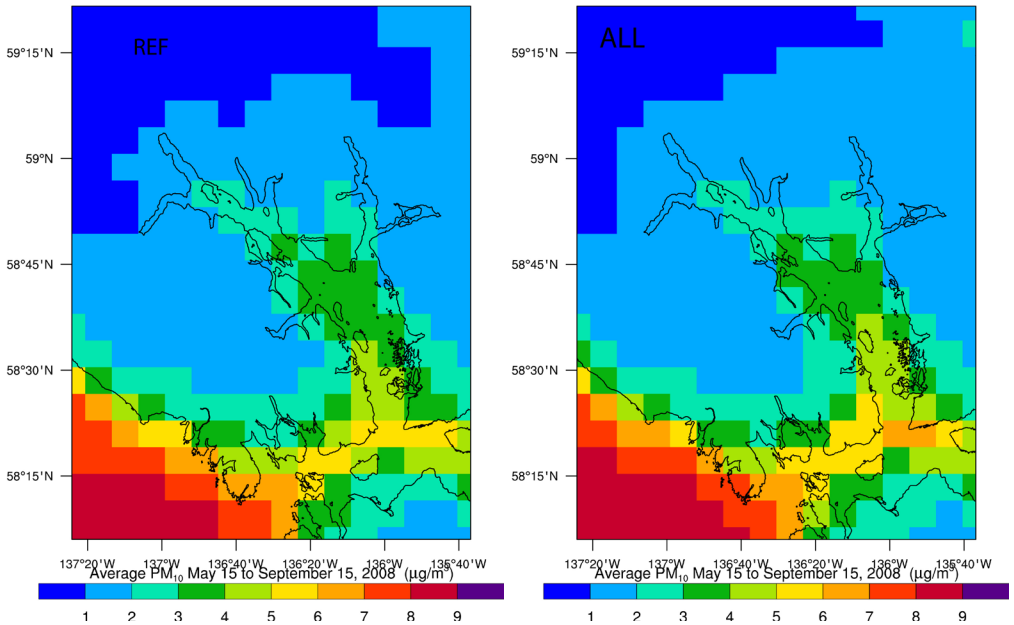


Figure 4. Season mean concentrations at cruise-ship height of (upper left to lower right) SO_2, NO_x, $PM_{2.5}$, and PM_{10}, in Glacier Bay National Park and adjacent areas as obtained by ALL and REF. Legends differ among panels.

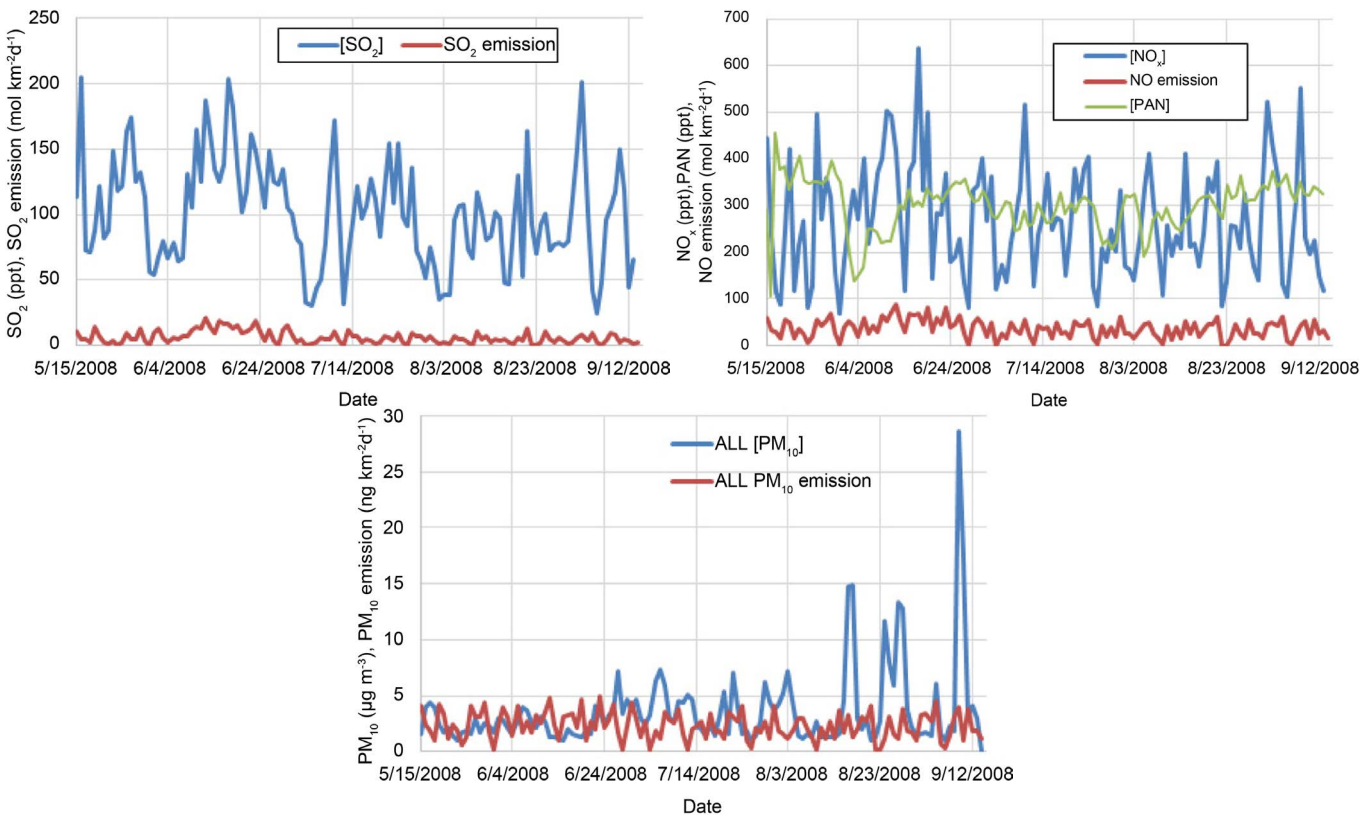


Figure 5. Bay-wide daily accumulated emissions of ALL SO_2, NO, and PM_{10}, and daily means of ALL SO_2, NO_x and PAN, and PM_{10} concentrations (upper left to lower middle) at cruise-ship height over the 2008 tourist season May 15 to September 15. Zero emissions indicate days without cruise-ship entries in Glacier Bay.

air composition over the water body (therefore not shown). On the contrary, cruise-ship emissions dominated the VOC concentrations over the water proper in REF [11] and ALL, as expected. Nevertheless, comparison of REF and ALL VOC concentrations confirmed that advection of VOC from anthropogenic sources outside of the bay had marginal impact on the VOC concentrations in the bay.

In Southeast Alaska, the complex terrain prohibits dairy and agricultural activities. Consequently, the major ammonia (NH_3) emission sources were anthropogenic. Highest NH_3 concentrations occurred in the maritime ABL over the shipping lanes and waterways as well as in the urban ABL of port cities due to ship emissions. Ammonia can neutralize nitric acid (HNO_3), which forms by NO_x oxidation, to produce ammonium nitrate aerosol (NH_4NO_3) [6] [48]. NH_3 can also neutralize sulfuric acid (H_2SO_4) by forming ammonium sulfate aerosol ($(NH_4)_2SO_4$). SO_2 oxidation forms H_2SO_4 under all atmospheric conditions in the presence of reactive radicals and water vapor [6] [41] [48]. Thus, ammonia served as a sink for oxidation products of SO_2 and NO_x and as a source for PM from precursor gases (section 3.3). These aerosols affect visibility [12].

3.2. Secondary Pollutants

In all four simulations, hourly O_3 concentrations were typically less than 50 ppb, sometimes even below 40 ppb in the maritime ABL of Glacier Bay. Thus, over the entire season, 8-hour mean near-surface O_3 concentrations remained below the EPA NAAQS of 70 ppb. Bay-wide season mean O_3 profiles differed negligibly between REF and ALL or CLN and ALL (**Figure 2**). Daily average O_3 concentrations remained less than 40 ppb in the first 200 to 500 m above the water surface and below 50 ppb up to about 2 km height (~800 hPa) most time. According to the O_3 concentrations obtained by RETRO and CLN, anthropogenic emissions occurring outside of Glacier Bay only marginally affected O_3 concentrations inside the bay. Comparison of REF and ALL-simulated O_3 concentrations confirmed this finding.

Over the entire season, ambient air temperatures governed PAN concentrations. Similar to O_3, anthropogenic emissions other than from cruise ships had small impact on the PAN concentrations found in Glacier Bay (therefore not shown). This finding indicates that besides temperature, cruise-ship emissions in the bay affected the PAN concentrations in Glacier Bay (**Figure 5**). The latter confirms [11], who reached this conclusion by comparing PAN concentrations of days with and without cruise-ship visits in Glacier Bay.

3.3. Primary and Secondary Aerosols

While cruise ships emit PM, PM also forms naturally from gas-to-particle conversion from precursor gases [6] [33] [41] [49]. As aforementioned, over Southeast Alaska, all PM was in the fine-particle mode with diameters less than 2.5 μm under undisturbed conditions (CLN). Consequently, the bay-wide season mean PM_{10} profile of CLN differed significantly from those obtained by REF, RETRO, and ALL with respect to concentrations (**Figure 2**), size distribution, and composition. This fact also means that all

PM_{10} in REF, RETRO, and ALL stemmed from anthropogenic emissions, gas-to-particle-conversion, and/or particle growth.

Over the season, in the first layers above surface, $PM_{2.5}$ emission-concentration relations behaved similar as found for SO_2 (**Figure 3**). Emission-concentration relations (correlation coefficients R up to 0.5) were significant over the international shipping lane, port cities, some water ways, and most of Canada. These findings indicate that residential sources, commercial ship traffic including cruise ships governed the near-surface PM concentrations in Southeast Alaska.

While locally in the bay under inversion conditions, $PM_{2.5}$ concentrations exceeded 35 $\mu g \cdot m^{-3}$ for several hours when one or two large cruise ships visited the bay on the same day, daily mean $PM_{2.5}$ concentrations remained below the 24-hour average of 35 $\mu g \cdot m^{-3}$ of the NAAQS [50] (therefore not shown).

Comparison of season bay-wide mean profiles (**Figure 2**) showed that sources other than cruise ships also contributed to the PM_{10} concentrations below 700 hPa (~3 km). In the case of PM_{10}, both advection of primary and secondary particles as well as gas-to-particle conversion from locally emitted and advected precursor gases (SO_2, NH_3, NO_x) played a role. Under strong inversion conditions, concentrations of PM increased strongly due to formation of secondary aerosols [50]. Consequently, concentrations of precursor gases like SO_2 and NO_x increased less (**Figure 5**) than expected from the emissions and/or by Equation (1) and were more sensitive to emissions and availability of oxidants.

In REF and ALL, cruise ships increased the concentrations of aerosol precursor gases in the bay (see **Figure 1**, **Figure 4**, **Figure 5** for SO_2, NO_x). Without cruise-ship emissions in Glacier Bay (CLN, RETRO) the PM distribution was nearly homogeneous over the bay (not shown, for CLN see **Figure 6** in [38]). On the contrary, in REF and ALL, enhanced PM concentrations occurred along the cruise path and close to the glaciers on season mean (**Figure 4**).

In the first decameters above the surface, PM_{10} concentrations were low over the ice fields of Glacier Bay National Park (**Figure 4**). This finding suggests that upslope transport of PM_{10} was low on season average. Indeed, the air in the ABL over the bay was decoupled from the air aloft frequently by inversions [9] [50].

3.4. Haze Index and Visibility

In all four simulations, days with worst visibility conditions were due to meteorology. At the height of cruise ships, bay-wide daily means of haze indices were high when relative humidity exceeded 90% (**Figure 6**). Daily bay-wide mean haze indices were correlated with relative humidity (R = 0.736, 0.618, 0.590, and 0.625 for CLN, REF, RETRO, and ALL, respectively).

CLN, REF, RETRO, and ALL daily bay-wide mean haze indices were also correlated with wind speed (R = 0.462, 0.825, 0.864, and 0.826, respectively). The high correlation coefficients found for REF, RETRO and ALL supported that transport of pollutants from emissions outside of Glacier Bay affected visibility inside the bay (**Figure 6**).

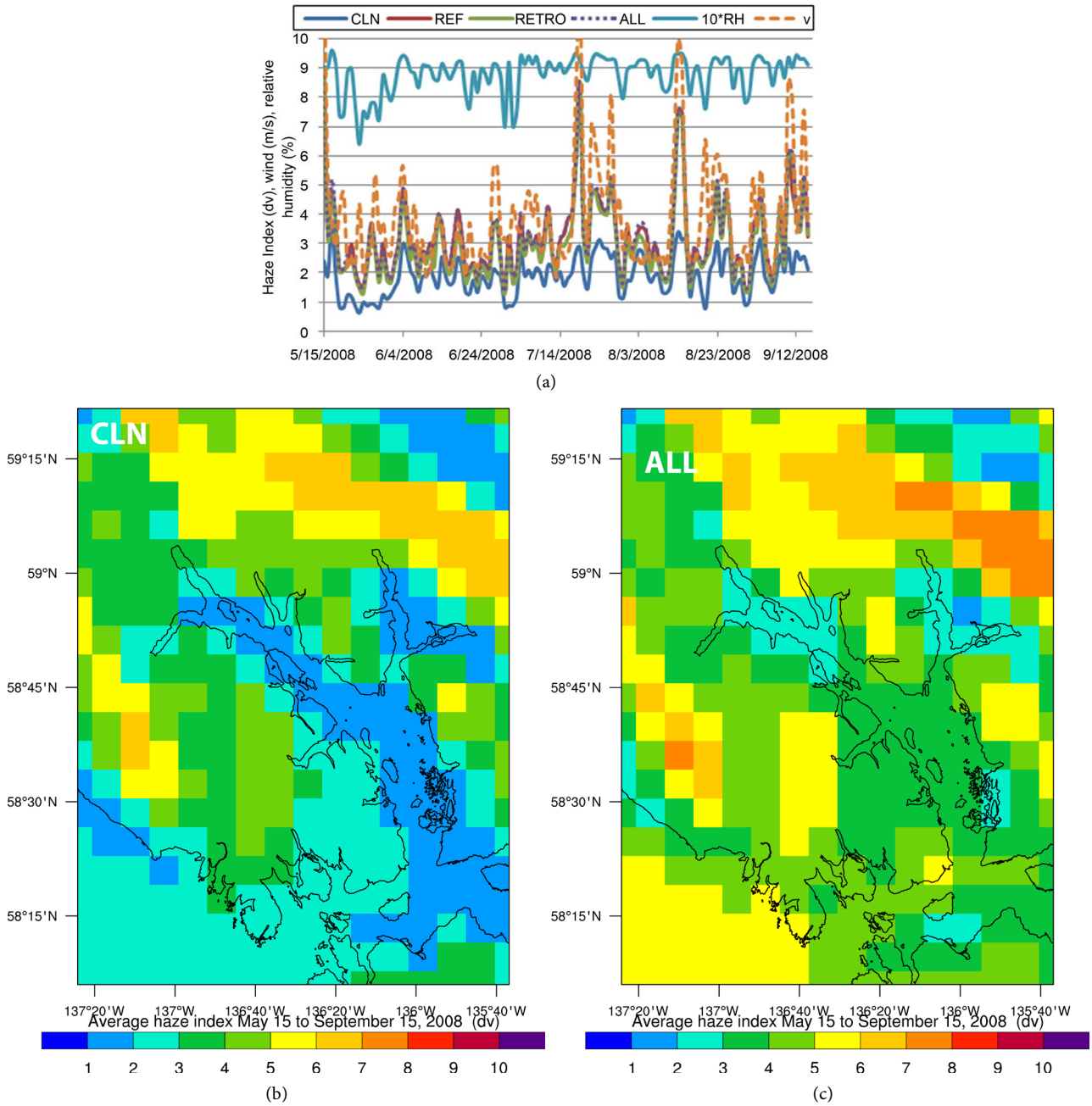


Figure 6. Comparison of haze indices at cruise-ship height (a) daily bay-wide average for the 2008 season, (b) in Glacier Bay as obtained by CLN (background conditions, no emissions except for activity-based biogenic emissions on season average), and (c) ALL (anthropogenic emissions including activity-based cruise ship and biogenic emissions on season average).

Daily bay-wide mean REF and ALL haze indices as well as RETRO and ALL haze indices were highly correlated (R = 0.991 and 0.995, respectively), while CLN haze indices were notably lower correlated with REF, RETRO, and ALL haze indices (R = 0.750, 0.730, and 0.756, respectively). The correlation of REF, RETRO and ALL haze indices with those of CLN confirmed [11] that meteorology governed visibility conditions in the bay. Correlations between REF and ALL haze indices and RETRO and ALL haze

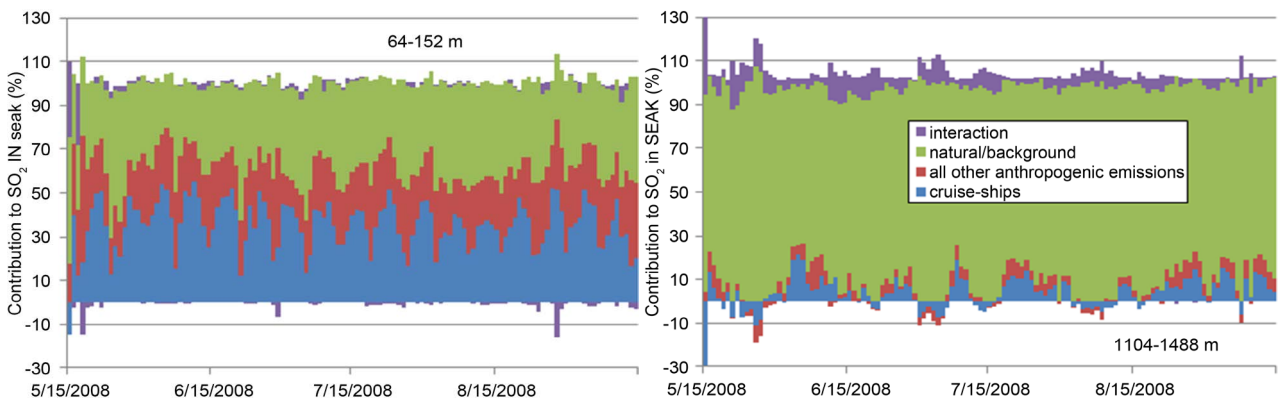
indices differed marginally. In RETRO, all ship emissions stemmed from commercial shipping, *i.e.* no cruise-ship emissions occurred in Southeast Alaska and Glacier Bay. Based on the low differences in correlations, we infer that transport of pollutants from ship emissions outside the bay affected the RETRO haze indices inside the bay.

In the bay, locally absolute differences between REF and ALL hourly haze indices reached up to 19.6 dv. Thus, transport of pollutants from emissions outside the bay influenced hourly visibility at least locally. Seasonal mean maximum (minimum) haze indices differed 2.0 (~0) between REF and ALL. These findings suggest that anthropogenic emissions occurring outside of Glacier Bay had small influences on worst visibility conditions that were caused by meteorological conditions.

Comparison of CLN and ALL haze indices revealed the total anthropogenic impact on visibility in Glacier Bay. On season average, the combined anthropogenic emissions increased the haze index by about 2 dv over wide areas of the bay compared to the natural conditions (Figure 6). Increases of about 1 dv or more occurred in ALL also in areas without cruise-ships traffic. These findings support that transport from outside and pollutants from emissions within the bay affected air quality, and visibility in the bay. Along the coast, in Icy Strait and Cross Sound, ALL anthropogenic emissions increased haze indices more than 3 dv on season average as compared to the background conditions represented by CLN. At these locations, cruise ships were the major emission source, but contributed only marginally to reduced visibility caused by meteorological conditions in the marine ABL.

3.5. Contributions of Emission Sources and Interaction

For all trace-gas species and PM, the ESCRs to the ALL concentrations differed in space and time (e.g. Figures 7-11). The ESCRs also differed among species partly due to different spatial occurrences of their emissions and/or emissions of their precursors (cf. Figure 1). Here we discuss contributions from the various emission sources to ALL concentrations by species. Discussion starts with the contributions to Southeast Alaska daily means at various heights followed by the seasonal mean contributions in Southeast Alaska to assess the degree to which air quality in Glacier Bay depends on emissions in the bay itself. Important indicators of potential for managing air quality inside



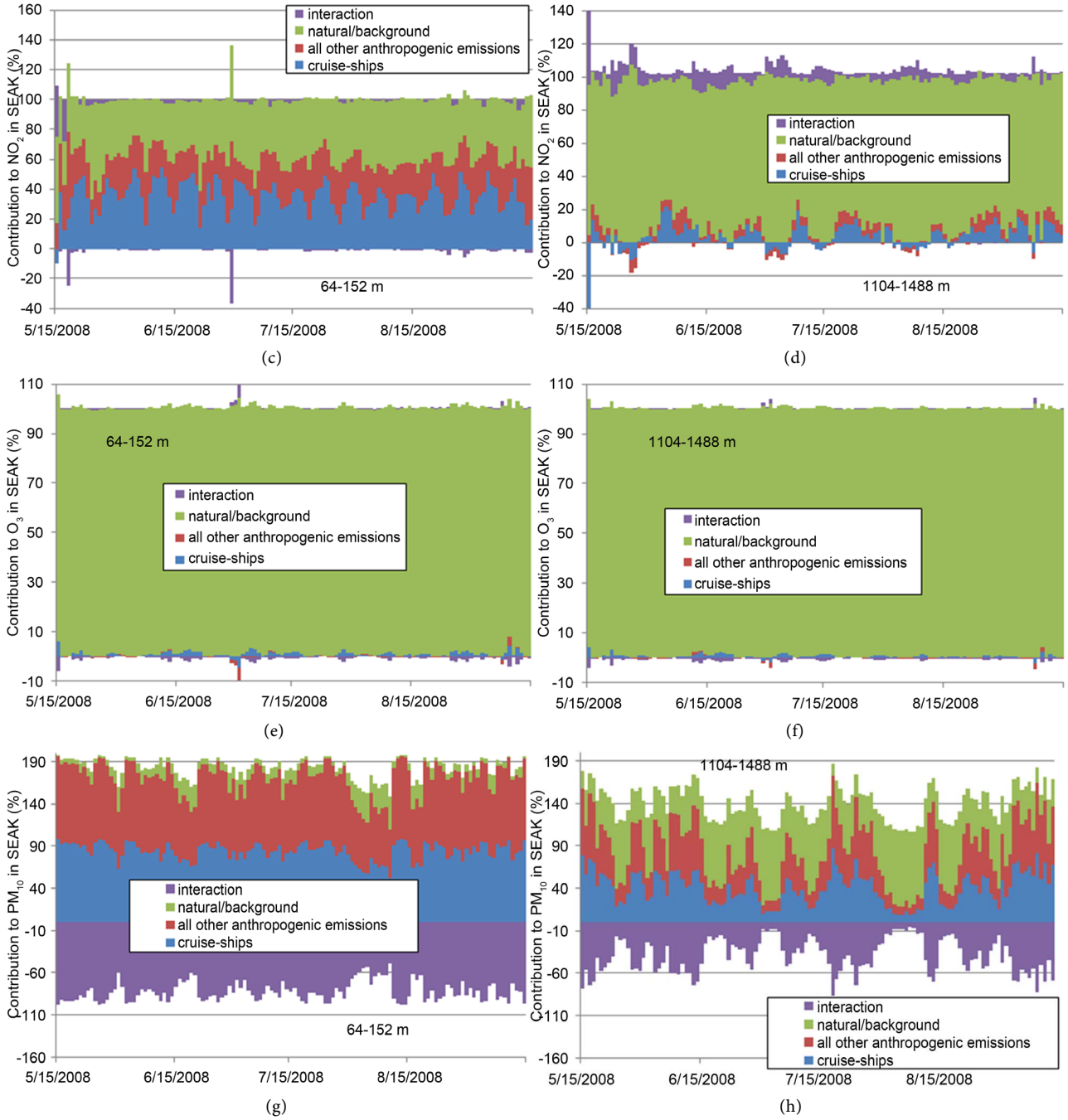


Figure 7. Daily mean ESCRs for natural emissions and background concentrations, cruise-ship emissions, all anthropogenic emissions except from cruise ships, and interaction of pollutants from these various sources to the Southeast Alaska mean ALL concentrations at layers between about (a) 64 - 152 m for SO_2, (b) 1104 - 1488 m for SO_2, (c) 64 - 152 m for NO_2, (d) 1104 - 1488 m for NO_2, (e) 64 - 152 m for O_3, (f) 1104 - 1488 m for O_3, (g) 64 - 152 m for PM_{10}, and (h) 1104 - 1488 m for PM_{10}. The layers between 64 - 152 m (left column) received the bulk of the cruise-ship emissions. The layers between 1104 - 1488 m (right column) represent the range of the top of the ABL that typically was around these heights over water. Chemical reactions among species emitted by different local and distant sources (interaction) can lead to both higher or lower concentrations in ALL than expected from superposition (Equation (2)). Negative ESCR values indicate that the respective emission source contributed to chemical reactions that reduced the concentration of the species in ALL as compared to when no such reactions would have occurred. Y-axes differ among panels.

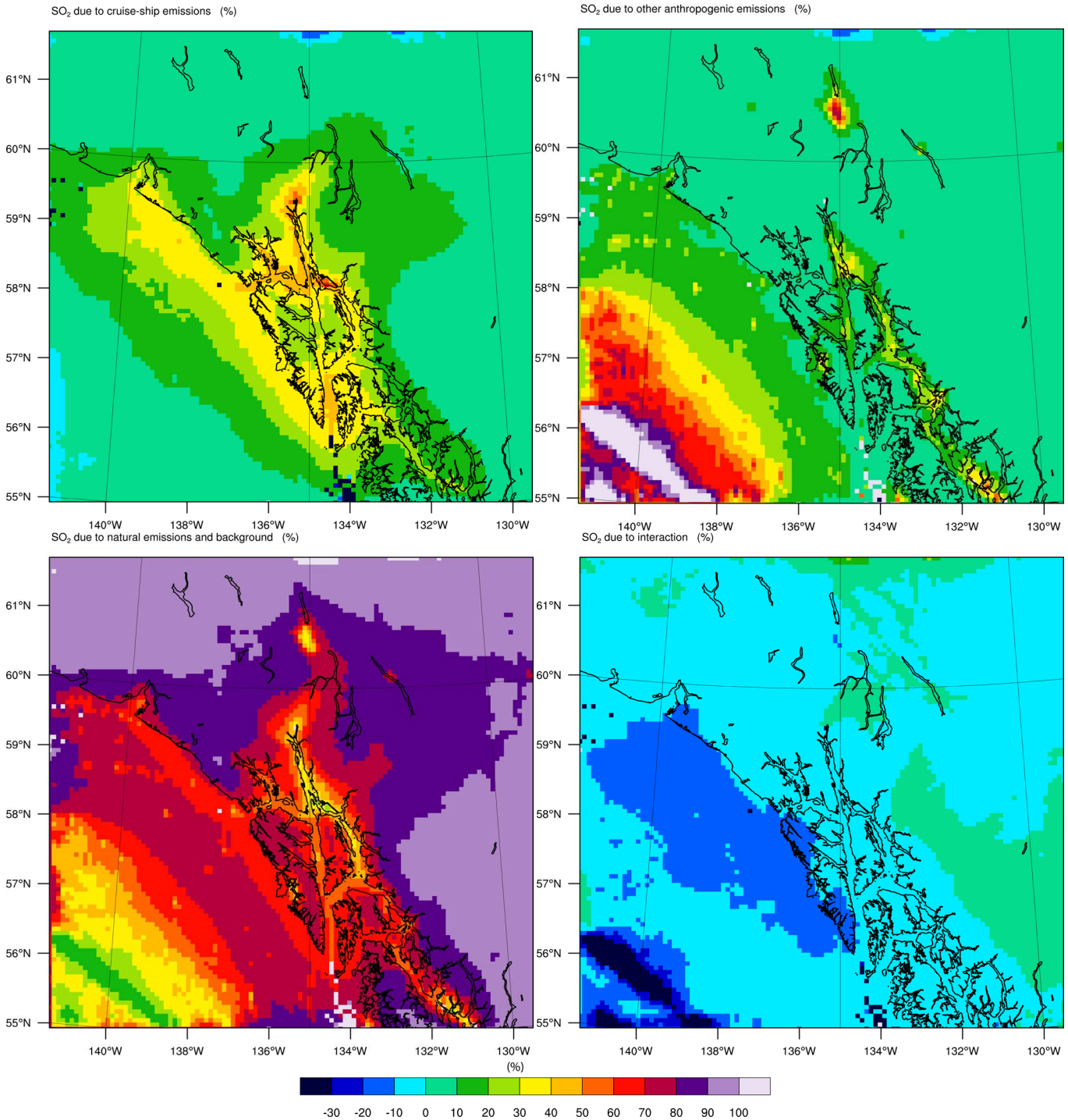


Figure 8. Season mean ESCRs (in percent) of (upper right to lower left) natural emissions and background concentrations ($ESCR_{CLN}$ = 100 × CLN/ALL), cruise-ship emissions ($ESCR_{REF}$ = 100 × (REF-CLN)/ALL), all anthropogenic emissions except from cruise ships ($ESCR_{RETRO}$ = 100 × (RETRO-CLN)/ALL), and interaction of pollutants (100 × α) from these sources to the ALL SO_2 concentrations at cruise-ship height in Southeast Alaska. Legend is valid for all panels.

the bay would be that the contribution from cruise-ships is highest in the bay, and the composition of pollutants inside and outside of the bay would differ.

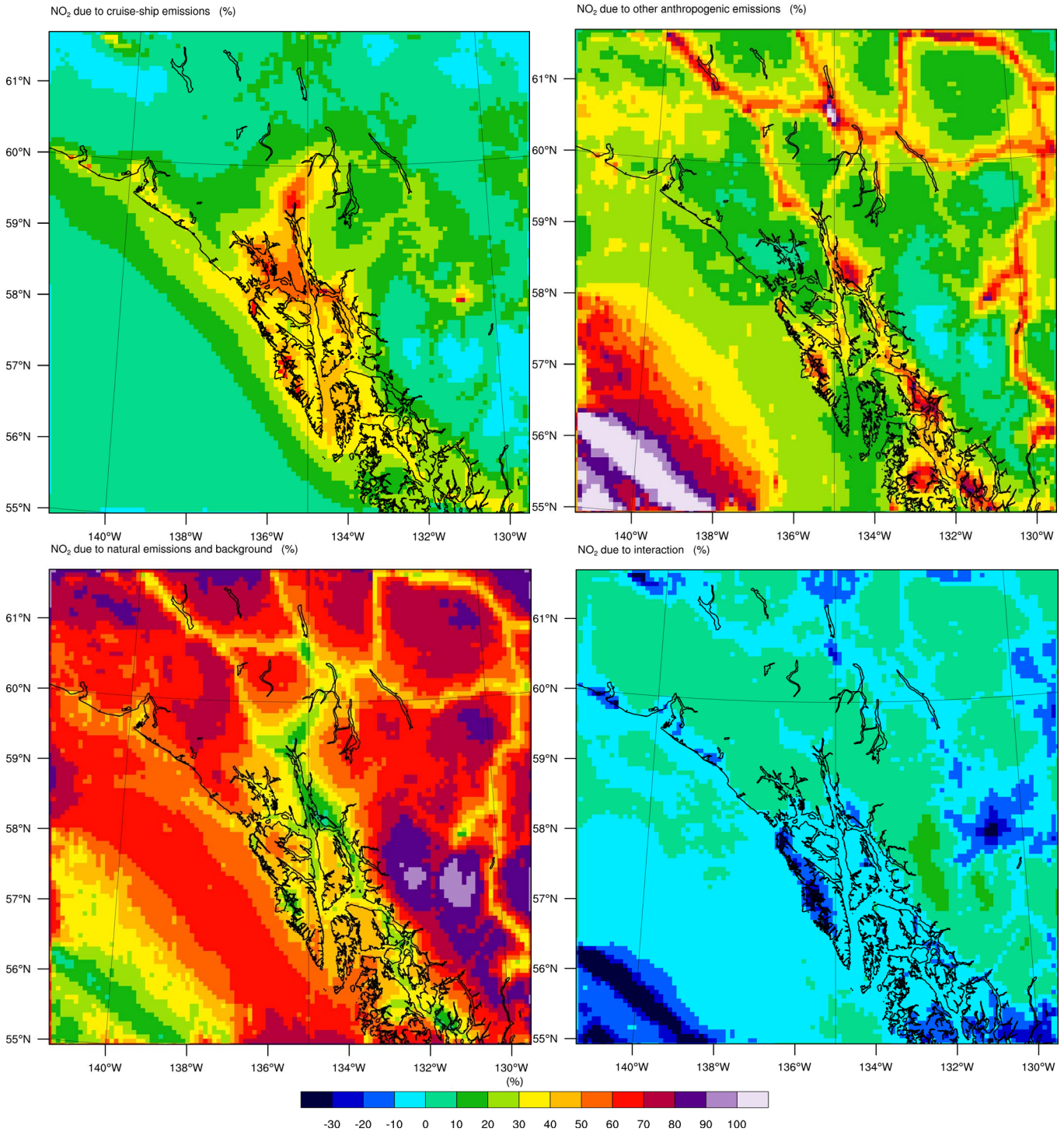


Figure 9. Season mean ESCRs (in percent) of (upper right to lower left) natural emissions and background concentrations ($ESCR_{CLN}$ = 100 × CLN/ALL), cruise-ship emissions ($ESCR_{REF}$ = 100 × (REF-CLN)/ALL), all anthropogenic emissions except from cruise ships ($ESCR_{RETRO}$ = 100 × (RETRO-CLN)/ALL), and interaction of pollutants (100 × α) from these sources to the ALL NO_2 concentrations at cruise-ship height in Southeast Alaska. Legend is valid for all panels.

3.5.1. Southeast Alaska Air Quality

On average over Southeast Alaska, in ALL, the majority of SO_2 in the lower ABL stemmed

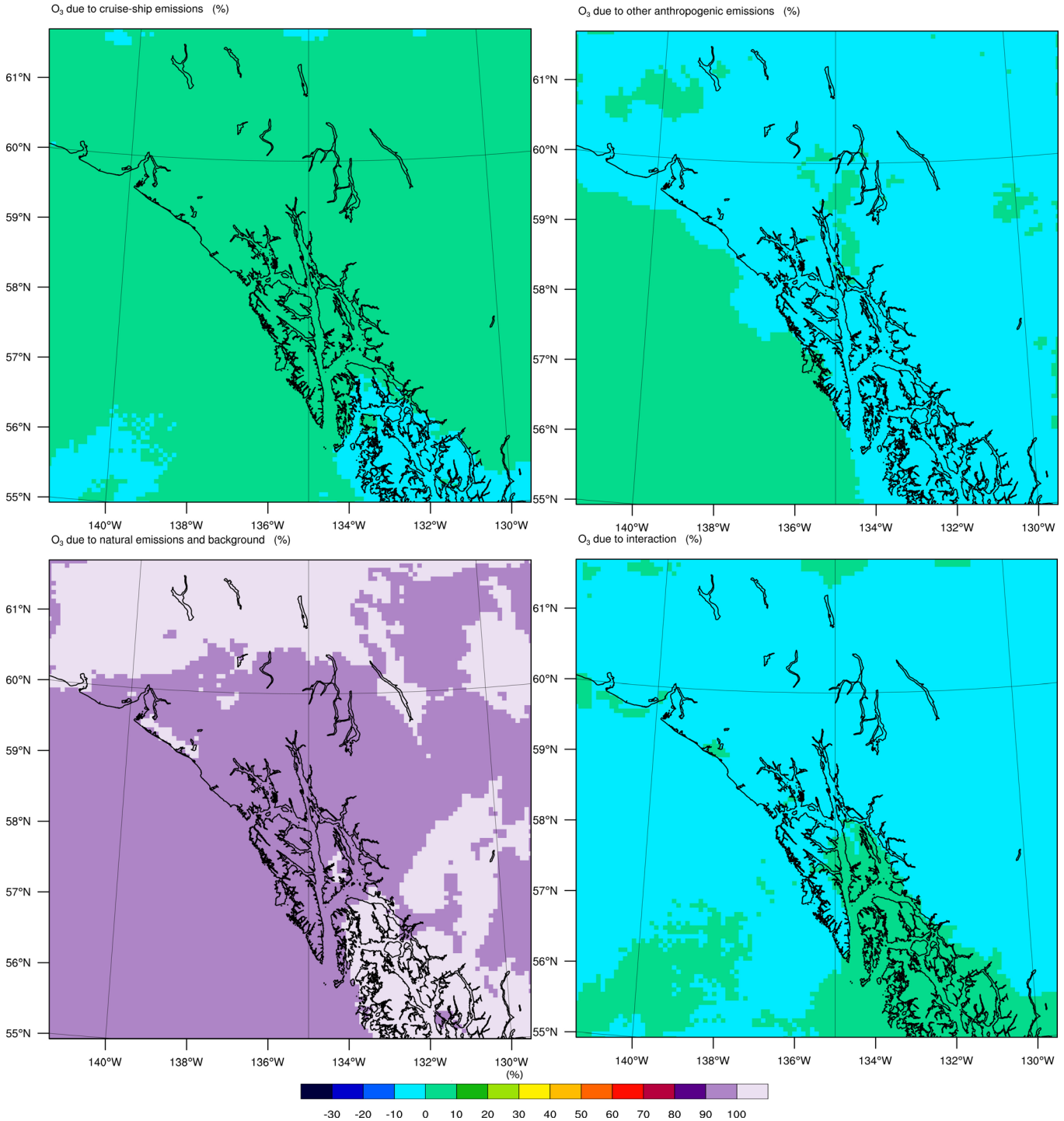


Figure 10. Season mean ESCRs (in percent) of (upper right to lower left) natural emissions and background concentrations ($ESCR_{CLN}$ = 100 × CLN/ALL), cruise-ship emissions ($ESCR_{REF}$ = 100 × (REF-CLN)/ALL), all anthropogenic emissions except from cruise ships ($ESCR_{RETRO}$ = 100 × (RETRO-CLN)/ALL), and interaction of pollutants (100 × *a*) from these sources to the ALL PM_{10} concentrations at cruise-ship height in Southeast Alaska. Legend is valid for all panels.

from cruise ship and anthropogenic emissions (e.g. Figure 7). At about 1.1 to 1.5 km height, cruise ships contributed up to 31% to the daily mean SO_2 concentrations in

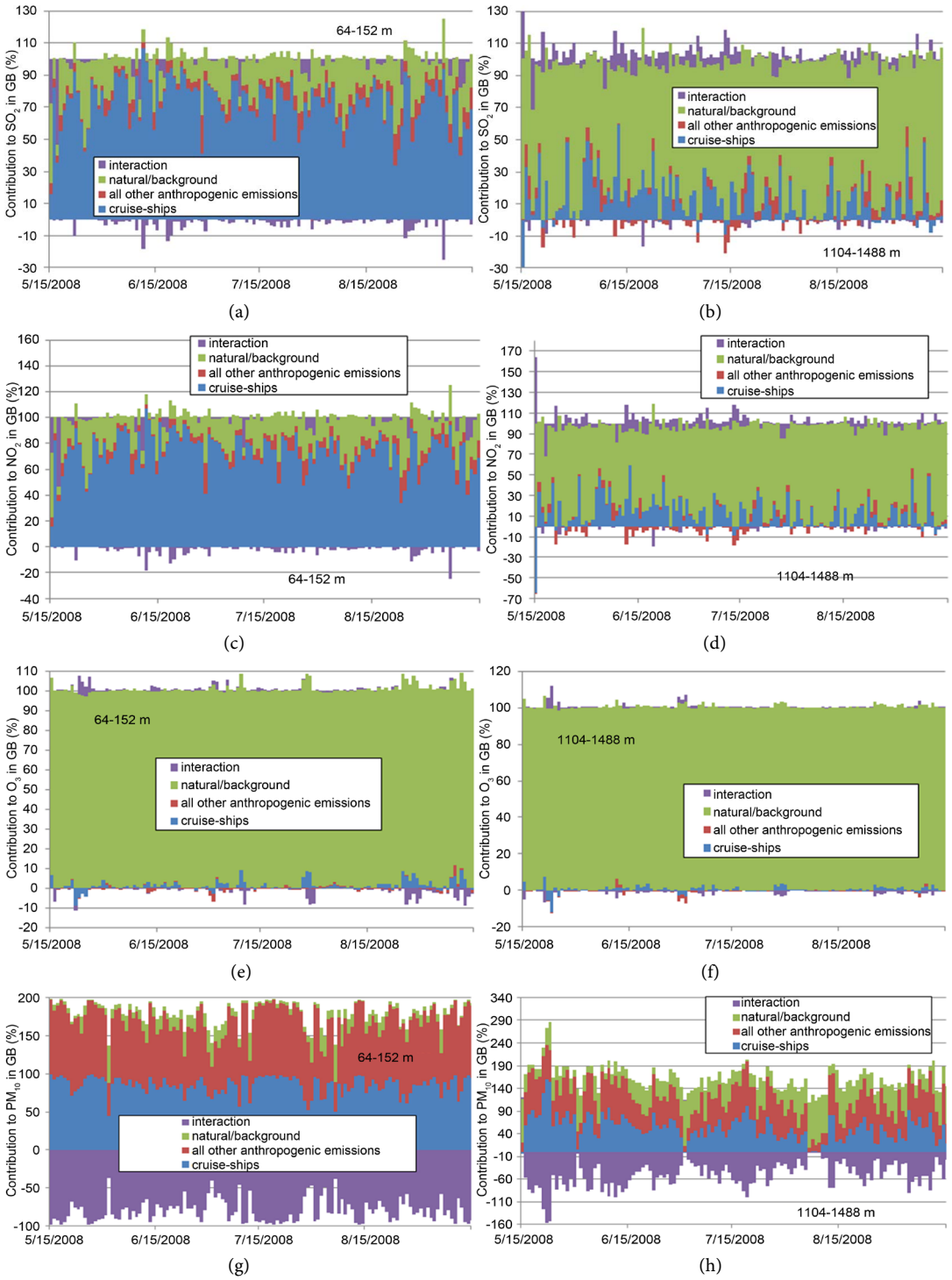


Figure 11. Daily mean ESCRs of natural emissions and background concentrations, cruise-ship emissions, all anthropogenic emissions except from cruise ships, and interaction of pollutants from these various sources to the bay-wide mean ALL concentrations at layers between about (a) 64 - 152 m for SO_2, (b) 1104 - 1488 m for SO_2, (c) 64 - 152 m for NO_2, (d) 1104 - 1488 m for NO_2, (e) 64 - 152 m for O_3, (f) 1104 - 1488 m for O_3, (g) 64 - 152 m for PM_{10}, and (h) 1104 - 1488 m for PM_{10}. The layers between 64 - 152 m (left column) are shown as these layers received the bulk of the cruise-ship emissions. The layers between 1104 - 1488 m (right column) are shown as the top of the ABL typically was below 1 km height. Note that y-axis legends differ among panels.

Southeast Alaska when several cruise ships were in the domain. Above the ABL, anthropogenic emissions contributed 10% or less to the daily mean SO_2 concentrations.

Often chemical processes among pollutants from various sources reduced the ALL SO_2 concentrations as compared to the concentrations expected from Equation (1). In the ABL, interaction of pollutants from different sources affected daily mean SO_2 concentrations less than 10% most time on Southeast Alaska average (Figure 7). However, occasionally up to 30% enhanced/reduced SO_2 concentrations occurred for short times (1 - 2 days).

At cruise-ship height, contributions from cruise ships remained below 30% in areas not travelled by cruise ships (Figure 8). In port cities, cruise ships contributed more than 60% to the season mean SO_2 concentrations. Cruise-ship emissions contributed 30% to 50% to the ALL SO_2 concentrations along the Alaska coast. In the waterways, contributions from cruise-ship emissions exceeded 50% in some locations. Over land, anthropogenic emissions from other sources than cruise ships contributed between 30% to 60% to the ALL SO_2 concentrations in urban areas. Commercial shipping contributed 100% to the ALL SO_2 emissions in the international shipping lane. However, more than 30% of the emitted SO_2 underwent chemical reactions and aerosol-formation processes thereby reducing the ALL SO_2 concentrations expected according to Equation (1). Consequently, on season mean, only up to 30% of the ALL SO_2 in the first 152 m or so over the international shipping lane were due to background concentrations (Figure 8).

On season average, the impact of interaction peaked between about 60 and 200 m or so in areas where cruise ships occurred. Recall that the bulk of cruise-ship emissions occurred between these heights. In the first decameters above the surface, ALL seasonal mean SO_2 concentrations were diminished by more than 4 and 5 ppt along the Alaska coast and over the waterways in the Alexander Archipelago, respectively, as compared to the values expected from Equation (1). Interaction reduced ALL seasonal mean SO_2 concentrations by up to 10 ppt over the international shipping lane (southwestern corner of the model domain). Interaction enhanced seasonal mean ALL SO_2 concentrations by more than 1 ppt over the southern part of the Coastal Mountains.

In the ABL, ALL daily and season Southeast Alaska mean NO_x concentrations behaved similar to those of SO_2 (Figures 7-9). At the top of the ABL, contributions by cruise-ship emissions to the SO_2 and NO_x concentrations in ALL were larger early in the season than in late summer and fall (Figure 7). The increased number of storms as summer progressed, fostered mixing and prohibited inversion formation and pollutant accumulation underneath.

In Southeast Alaska, anthropogenic emissions other than from cruise ships and cruise-ship emissions contributed up to 40% and 30% to the daily regional mean NO_2 concentrations (Figure 7) below 1 km height. Non-linear interaction of pollutants contributed less than ±10% to the NO_2 concentrations mainly below 1 km in the ABL.

Interaction of trace gases from other sources with NO_x reduced ALL NO_x concentrations by more than 10% over land along most of the Gulf of Alaska Coast, the interna-

tional shipping lane, and the crossings of major highways (Figure 9). Around 150 m height over Sitka, reduction ranged still between 20 and 40 ppt. The magnitude of diminution decreased with height to about 4 to 8 ppt for this port city. At Skagway, interaction of pollutants from all sources enhanced NO_x concentration by up to 20 ppt above 152 m or so as compared to the values expected from Equation (1). Interaction of pollutants increased with increasing height to around 400 m height and then decreased again. Between about 250 and 400 m height, NO_x enhancement of this magnitude also occurred along some waterways, the Coastal Mountains, and coastal port cities. Between 600 and 1100 m, ALL NO_x concentrations were 4 to 6 ppt higher than expected by Equation (1). Typically, interaction became marginal over these areas above the top of the ABL between 2.3 and 2.9 km height (therefore not shown).

However, over Canada in the lee of the Coastal Mountains, interaction enhanced ALL NO_x concentrations locally by up to 3 ppt at heights between 1.9 and 2.9 km (not shown). Here, high reaching convection transported pollutants upward and out of the ABL. On the contrary, over water, the comparatively higher stability than over land restricted vertical mixing and exchange with aloft air. Consequently, interaction of pollutants from different sources was restricted to a lower height over water than land.

In some areas of Southeast Alaska, ALL NH_3 concentrations were diminished by up to 10 ppt as compared to the values expected from Equation (1). In the first decameters over Sitka, for instance, NH_3 interacted with reactive gases and aerosols from the various sources diminishing ALL NH_3 concentrations by up to 10 ppt. Slight diminutions also occurred in the first decameters over some tidal glacier fjords outside Glacier Bay. Above that height, no significant interaction of pollutants from various sources with NH_3 occurred.

In ALL, like in the other simulations, background O_3 dominated the O_3 concentrations (Figure 10). Cruise ship and other anthropogenic emissions marginally affected the daily mean Southeast Alaska O_3 concentrations. In the ABL, cruise ships contributed less than 5% on Southeast Alaska average to the ALL O_3 concentrations (Figure 7).

In the first decameters above ground level, interaction of pollutants from the various emission sources increased ALL O_3 concentrations up to 0.6 ppb over the southern part of the Alexander Archipelago. On the contrary, ALL O_3 concentrations were diminished up to 0.9 ppb over Canada in the northeastern part of the model domain (not shown). The impact of interaction on ALL daily mean O_3 concentrations was highest in the layers that received the bulk of cruise-ship emissions in areas where cruise ships were present (not shown). Above that height, the absolute magnitude of interaction decreased slightly with increasing height in these regions and Southeast Alaska wide (e.g. Figure 7). Overall, interaction of pollutants from different sources and locations had negligible impacts on Southeast Alaska O_3 concentrations.

On average, PAN depletion due to interaction increased up to 40 ppt landwards below 1 km above ground. Above this height, the general pattern remained, but with two orders reduced magnitude as compared to the layers below 1 km.

In Southeast Alaska, under clean background conditions (CLN), no PM_{10} with diameters greater than 2.5 μm occurred, *i.e.* all PM_{10} was $PM_{2.5}$. Consequently, in ALL, PM_{10} exceeding 2.5 μm in diameter (coarse particles) stemmed from cruise ship and anthropogenic emissions, and/or particle growth by gas-to-particle conversion from precursor gases. In ALL, less than 10% of PM_{10} was $PM_{2.5}$ on most days.

Interaction between the different anthropogenic and cruise-ship emissions affected PM type, size, and sedimentation. It diminished PM_{10} concentrations in the first decameters along the coast and in waterways (not shown). On season average, maximum diminution occurred west of Cross Sound (>8 $\mu g \cdot m^{-3}$). Interaction between pollutants from cruise-ship emissions and other sources decreased with height (**Figure 7**). The pattern remained broadly the same with increasing height, but the horizontal extension of areas with interaction increased slightly due to atmospheric transport (not shown). Above 1.5 to 2 km, interaction became marginal. Interaction diminished the season average ALL PM_{10} concentrations up to 8 $\mu g \cdot m^{-3}$ over the Pacific Ocean over the international shipping lane.

3.5.2. Glacier Bay

In Glacier Bay, ESCRs for the ALL seasonal and daily means of SO_2, NO_x, and PM_{10} concentrations showed distinct differences compared to those over Southeast Alaska (**Figure 7**, **Figure 11**). Typically, cruise-ship emissions showed a greater percentage contribution to the bay-wide daily mean ALL concentrations than to the Southeast Alaska wide ALL concentrations.

In Glacier Bay, cruise-ship emissions contributed up to about 80% of the daily mean SO_2 concentrations below 1 km on days with cruise-ship visits (**Figure 11**) and between 40% and 70% to the bay-wide daily mean NO_2 concentrations below 1 km height. Similar to Southeast Alaska, non-linear interaction of pollutants contributed less than ±10% to the NO_2 concentrations and mainly below 1 km in the ABL (**Figure 7**, **Figure 11**). Anthropogenic sources others than cruise ships marginally contributed to the ALL SO_2 and NO_2 concentrations in the layers into which the cruise ships emitted.

In Glacier Bay, highest contributions by cruise ships to the ALL NO_2 concentrations occurred early in the season (**Figure 11**). At about 250 to 400 m, interaction enhanced ALL NO_2 concentrations by 4 to 8 ppt close to the glaciers as compared to the values expected from Equation (1). Above 2 km, ALL daily mean SO_2 and NO_2 concentrations in Glacier Bay typically represented the natural background conditions (not shown).

In Glacier Bay, anthropogenic sources other than cruise ships contributed to the SO_2 and NO_x concentrations marginally in the layers into which the cruise-ships emitted (e.g. **Figure 9**). These contributions rarely exceeded 10% and were due to advection of pollutants from emissions outside of the bay. In the ABL over the bay, cruise-ship emissions contributed about 60% and up to 80% or more to the bay-wide daily mean SO_2 and NO_x concentrations. Even on days without cruise-ship visits, cruise-ship emissions from previous visits still made up for more than 40% of the bay-wide daily means of ALL SO_2 and NO_x concentrations in layers into which cruise ships emitted. At the top of the ABL, background concentrations governed the mean SO_2 and NO_x concen-

trations over the bay on most days.

For O_3 concentrations, cruise-ship emissions and advection of pollutants from other anthropogenic sources contributed less than 10% on average to the bay-wide daily mean (**Figure 11**). In the bay, natural background O_3 concentrations made up more than 90% of the total ozone.

The lower mean contribution of natural sources and background concentrations to PM over the bay (**Figure 11**) than Southeast Alaska (**Figure 7**) resulted from the low $PM_{2.5}$ emissions from sea-spray. On season average, winds were calm in the bay [9] [38].

The ESCRs indicated that advection of PM from outside of the bay was small most of the time. In Glacier Bay, interaction among pollutants from cruise ships and other sources diminished PM_{10} concentrations up to 2 $\mu g \cdot m^{-3}$ on season average (not shown). As expected, in Glacier Bay, interaction was strongest where cruise ships berthed in the layers receiving the bulk of the cruise-ship emissions. Lowest interaction occurred in areas without cruise-ship travel. Note that in the former and latter areas, diminution amounted up to 3.3 and 0.7 $\mu g \cdot m^{-3}$, respectively.

In Glacier Bay, interaction between pollutants from cruise-ship emissions and sources outside of the bay decreased with height for all species examined (**Figure 11**). It vanished between 800 and 1100 m. This blending height was much lower than the daily averages of 1100 to 1400 m found over Southeast Alaska (cf. **Figure 7**, **Figure 11**). The different height was due to the higher SST in the Pacific Ocean and the higher surface temperatures over non-ice covered land in most of Southeast Alaska than in Glacier Bay. The frequent inversions in Glacier Bay also contributed to the different height at which concentrations represented the background concentrations.

Due to thermodynamic reasons (Köhler curve), large water-soluble particles swell at lower relative humidity than the small ones [6]. The size of the particles, however, affects visibility [51] and explains the about 2 dv mean reduction in visibility in ALL as compared to CLN (**Figure 6**).

4. Conclusions

The limits of managing air quality are set by the contribution of the emission sources under control to the total concentrations of the species. To assess the limits to which the National Park Service (NPS) can manage air quality within Glacier Bay we setup four WRF/Chem simulations that permitted calculation of emission-source contribution ratios (ESCRs). These simulations were performed over the length of the 2008 peak tourist season (May 15 to September 15). They only differed by the type of emissions considered 1) only biogenic emissions (CLN), 2) biogenic and cruise-ship emissions (REF), 3) biogenic and anthropogenic emissions except cruise-ship emissions (RETRO), and 4) biogenic and anthropogenic emissions including cruise-ship emissions (ALL). In this study, ALL represented the actual atmospheric composition over Southeast Alaska and Glacier Bay. Focus was on primary and secondary pollutants as well as particles related to cruise-ship emissions as the NPS can control the speed, number of entrances,

among other things in Glacier Bay.

In general, in Southeast Alaska, the ESCRs to the ALL concentrations differed among species partly due to the spatial-temporal variability of their emission sources as well as meteorological conditions. For all species examined, interaction between pollutants from cruise-ship emissions and other anthropogenic sources decreased with height. Interaction became negligible above the top of the ABL except where convection transported pollutants into the free atmosphere.

Local sources governed air-quality in Southeast Alaska. Local emissions dominated the concentrations in the ABL around area sources like settlements including port cities, along line emission sources like waterways, shipping lanes, and major highways. In case of Glacier Bay, local sources were cruise ships and biogenic emissions.

In Southeast Alaska, all coarse PM (particles with diameters > 2.5, but ≤10 μm) was due to anthropogenic sources including cruise ships. Residential sources and cruise-ship emissions governed SO_2 and PM_{10} concentrations in the coastal ABL of Southeast Alaska. Cruise-ship emissions contributed 30% to 50% to the ALL SO_2 concentrations along the Alaska coast. Commercial shipping other than cruise ships governed the SO_2 and PM_{10} concentrations in the maritime ABL over the international shipping lane. Here only 30% of the ALL SO_2 was due to the natural background concentrations. In port cities, cruise ships were the main cause for the ALL SO_2, NO_x, NH_3, and PM_{10} concentrations. In Canada, road traffic was the main contributor to the ALL NO_x concentrations in the ABL. Non-linear interaction of pollutants from various emission sources contributed on average less than ±10% to the ALL NO_2 concentrations in Southeast Alaska.

Below the ABL, the contributions of the various emission sources to daily mean concentrations of ALL SO_2, NO_x, and PM_{10} showed distinct differences between Southeast Alaska-wide and Glacier Bay bay-wide daily means.

In general, in Glacier Bay, the percent contribution of cruise-ship emissions to the concentrations varied with meteorological conditions. Highest percentage contributions from cruise ships occurred early in the season when inversions occurred more often than at the end of the season. Later in summer and fall, the number of cyclones increased vertical mixing with clean air from aloft thereby diluting the pollutants in the ABL.

In Glacier Bay, cruise-ship emissions typically contributed between 60% and 80% to the bay-wide daily mean NO_2 concentrations below 1 km height on days with visits. On days without cruise-ship entrances, emissions from previous cruise-ships entrances explained 40% of the bay-wide daily mean ALL SO_2 and NO_x concentrations. Anthropogenic emissions other than those from cruise ships rarely contributed more than 10% to the daily mean PM_{10}, SO_2, and NO_x concentrations in the ABL of Glacier Bay. Together these findings mean that air quality in Glacier Bay was governed by cruise-ship emissions most of the time. Thus, we conclude that there is potential for managing air quality at these times.

In Glacier Bay, for all contaminants, interaction was lowest in the areas without

cruise-ship traffic and highest where cruise ships berthed for glacier viewing. Here, interaction enhanced the ALL NO_2 concentrations by 4 to 8 ppt at heights between about 250 and 400 m. Interaction of precursor gases and particles from various sources diminished PM_{10} concentrations up to 2 $\mu g \cdot m^{-3}$ on season average. Like in Southeast Alaska, less than ±10% of the ALL NO_2 concentrations below 1 km was due interaction of pollutants from various sources. Over the bay, interaction typically became marginal between 800 and 1100 m, which was typically about the height of the ABL. Above the ABL, background concentrations governed the mean SO_2, NO_x, NH_3, VOC, and PM_{10} concentrations over the bay on most days.

Together, the findings suggest that the NPS may be able to effectively manage air quality within Glacier Bay at least on days with stagnant air conditions. Then, the air in the bay is nearly cut off from advection of pollutants from other sources and cruise-ship emissions, *i.e.* pollutants from local controllable sources, govern the magnitude and distribution of daily mean ALL SO_2, NO_x, NH_3, and PM_{10} concentrations in the bay. During most other weather conditions, pollutants from other sources advected into the bay limited the margin to which the local cruise-ship emissions contribute to the overall concentrations. Since the NPS can impose emission-control measures (e.g. speed limits, low sulfur fuel, etc.), we conclude that the highest potential for managing air quality and hence visibility is under stagnant conditions in Glacier Bay.

We also conclude that managing local emissions does not necessarily equate to managing visibility because set emissions may or may not equate to haze and/or reduced visibility depending upon atmospheric composition and weather conditions. In Glacier Bay, worst visibility days were caused almost exclusively by meteorology. On season average, the combined anthropogenic emissions increased the haze index only by about 2 dv over wide areas of the bay as compared to the natural conditions. In fact, increases of about 1 dv or more occurred in ALL in areas of the bay that had no cruise-ship traffic, such as the eastern arm. This result suggests that pollutants from cruise-ship emissions in the bay can cause about the same degradation of visibility as pollutants advected from outside the bay. Consequently, we conclude that emission-control measures do not lead automatically to improved visibility.

The results showed that all coarse particles were due to anthropogenic emissions. Due to thermodynamic reasons (Köhler curve), coarse water-soluble particles swell at lower relative humidity than the fine ones. Coarse particles reduce visibility and explain some of the 2 dv mean reduction in visibility in ALL as compared to CLN. Thus, demanding filters or scrubbers to reduce the emissions of coarse particles could delay the onset of swelling towards comparatively higher relative humidity and finer particles than required for swelling of coarse particles.

Reducing NO_x emissions and/or use of low sulfur fuel would be indirect emission-control measures that target precursor gases of particle formation. Since gas-to-particle conversion takes time, such measures may be only beneficial during long stagnant conditions when the air remains in the bay and pollutants accumulate underneath inversions. However, such implementation would require forecasting long stagnant condi-

tions several days ahead of time to maximize the effect.

Finally, we caution that our results focus on air quality from the perspective of daily or season average air quality and visibility conditions, *i.e.* they are not based on specific management goals. Such goals may include thresholds when haze is produced due to ship traffic. We also caution that the deposition of pollutants can affect or even alter park ecosystems if they accumulate in organisms and/or alter the pH-value of water, snow and soil water. Particle accumulations on glaciers, particularly in the form of black carbon, can also affect the radiation budget and local climate. Thus, while we conclude that the air quality is generally high in Glacier Bay, and propensity for haze production and thus reduced visibility typically occurs only under certain conditions such as strong inversions and/or high relative humidity, the impacts of pollutants from cruise-ship emissions can occur in a myriad of other ways that were beyond the scope of the study.

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