EXPLORING EFFECTS OF AEROSOL PROPERTIES ON PHOTOLYSIS RATE CALCULATIONS FOR NASA'S POLCUBE CUBESAT POLARIMETER

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Abstract

The Fast-J model calculates photolysis rates (J-Values) in an atmosphere with an arbitrary number of aerosol layers. These aerosol light interactions can change the local photochemistry in the planetary boundary layer. Understanding photochemical changes at the surface is critical for accurate air quality forecasting. Fast-J calculations with pre-determined aerosol layer heights have resulted in differing J-Values at the surface by as much as 50%. To further explore the effects of aerosol layer heights on J-Values, we turn to NASA's Aerosol Cloud meTeorology Interactions oVer the western ATlantic Experiment (ACTIVATE). From ACTIVATE, aerosol layer heights are calculated using the NASA High Spectral Resolution Lidar 2 (HSRL-2) and Research Scanning Polarimeter (RSP). This work is part of an effort to create a J-Value product for the upcoming NASA PolCube mission. The absence of a co-located lidar on PolCube complicates validation of aerosol layer height retrievals. ACTIVATE provides coincident lidar and polarimeter measurements, making it ideal to investigate how uncertainties in polarimetric aerosol layer height retrievals affect J-Values. Insights from this analysis will guide the development of a PolCube aerosol layer height and tropospheric J-Value product, as part of my work funded by the Virginia Space Grant Consortium graduate research fellowship.

Introduction

With more frequent wildfires across the continent bringing smoke, as well as the steady supply of sea salts from the Chesapeake Bay and carbon from the coal piles near I-664 in Newport News, the Hampton Roads area has no shortage of potential for containing atmospheric aerosols. Aerosols are small particles, ranging from nanometers to tens of microns, that get suspended into the atmosphere[2,3]. Smoke and soot are examples of small aerosols, usually named PM2.5 (particulate matter under 2.5 microns), while sea salts are a larger example and named PM10 (size below 10 microns).

Aerosol interactions with radiation have both climate and health effects[2]. Aerosols still yield the largest uncertainties in the radiation budget of Earth, making quantifying their long-term effects challenging[6,7]. Cloud radiative effects are next in terms of high uncertainty in the radiation budget. Clouds also have the ability to interact and have their properties changed by aerosols, increasing aerosols' importance in Earth's radiation budget. Aerosols can cause respiratory damage when breathed in directly, but they can also interact with radiation and other species that create secondary products that also cause damage when inhaled[4,5]. When a layer of aerosols are present in the atmosphere, they can absorb and scatter sunlight, blocking light from traveling farther down as it is added to the light above the layer. This can change the local photochemistry observed around the aerosol layer. A photochemical reaction is a chemical reaction where the energy needed to react is deposited by a photon. An example is the destruction of ozone. Ozone destruction usually happens high in the stratosphere; however, if there is a concentration of ozone near the surface, an aerosol layer can limit the amount of radiation at the surface to destroy the ozone. Stratospheric ozone is vital to human health by protecting the surface from high-energy UV photons, but can be harmful if breathed in at the surface[5]. These byproducts of aerosols' interaction with radiation have led to this area of study being labeled as important by the National Research Council Decadal Survey for Earth Science and Applications from Space[1].

This study uses the Fast-J model to explore how photolysis rates can be affected by aerosol properties. Fast-J is an algorithm that calculates photolysis rates through the entire atmospheric column for an atmosphere with arbitrary aerosol and cloud layers[7,8]. Photolysis rates can be used in global chemical models to forecast air quality and gas species abundances in the atmosphere[9]. Important aerosol properties the model needs are aerosol optical depth (AOD), single scattering albedo (SSA), aerosol layer height, etc.. This study will focus on how the aerosol layer height affects photolysis rates. Figure 1 shows how differences in the (urban pollution type) aerosol layer height can result in different photolysis rates around the layer and at the surface for the destruction of NO2.

With arbitrarily picked aerosol layer heights, Fast-J shows large differences in surface photolysis rates. To quantify the sensitivity of photolysis rates to aerosol layer height we utalize data from NASA's Aerosol Cloud meTeorology Interactions oVer the western ATlantic Experiment (ACTIVATE) mission. The ACTIVATE campaign flew out of NASA's Langley Research Center (LaRC) from the winter of 2020 to the summer of 2022 for a total of 179 flights[15]. Throughout the flight campaign, two aircraft were flown to take observations. The first is NASA's HU-25 Falcon, which is specialized for in situ trace gas, aerosol, and cloud variables. The next is NASA's B200 King Air that has the capability of remote sensing observations of aerosols and clouds, along with dropsondes for meteorological variables[15].

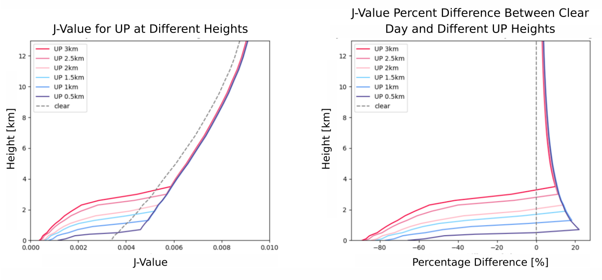

Figure 1: Left: Plot of photolysis rate profiles for a clear day and atmospheres with ascending layers of urban pollutant aerosols from 0.5-3km. Right: Plot of the profiles of the photolysis rate percent difference of each layered case to a clear day.

Aboard the flights were two instruments of interest for what is needed to run Fast-J. First, general aerosol properties such as type, size, and SSA are retrieved from the Research Scanning Polarimeter (RSP). Polarimetry allows for a passive retrieval of aerosol properties, and is not limited to a single line of sight like other remote sensing methods. The second is the second-generation High Spectral Resolution Lidar (HSRL-2). The trade-off of only retrieving a line-of-sight measurement is a more precise layer top retrieval. Both RSP and HSRL-2 have their own aerosol layer height retrievals. To explore the changes in photolysis rates due to aerosol layer height, heights retrieved from both HSRL-2 and RSP are used as inputs for Fast-J, with HSRL-2 heights labeled as ”truth” heights.

This work is done with the future goal of creating a photolysis rate pipeline for NASA's PolCube CubeSat. PolCube is an orbiting polarimeter scheduled for launch in 2026 to study aerosols with higher accuracy[11]. PolCube has two separate detectors separated by a viewing angle of 50 degrees and has a wider wave-

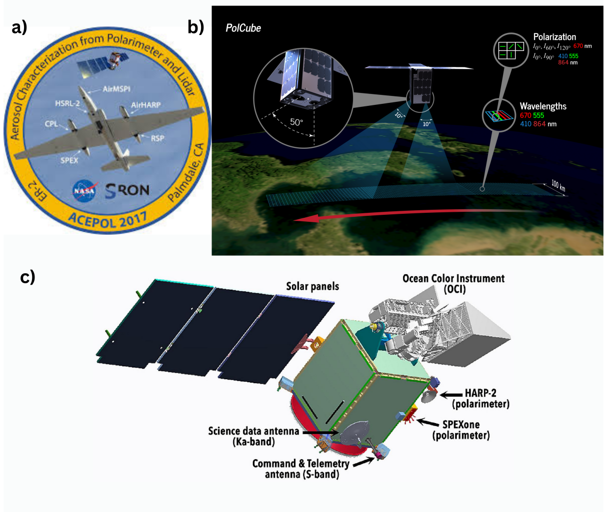

Figure 2: a) ACTIVATE mission patch showing the B200 King Air housing the RSP and HSRL-2 instruments. b) Graphic of PolCube CubeSat polarimeter with wavelengths, polarization states, and swath path shown. c) PACE satellite with labeled instruments including the HARP-2 and SPEXone polarimeters.

length range than RSP (farther into the UV), allowing for more accurate property retrieval. Since PolCube is not operational yet, we will use NASA's Plankton, Aerosol, Cloud, Ocean Ecosystem (PACE) satellite[16]. PACE has multiple polarimeters on board and has been releasing data since 2024. We use PACE to create a photolysis rate pipeline as a proof of concept for a pipeline for PolCube.

The analysis is conducted in two separate parts with the goal of creating a photolysis rate product pipeline for PolCube. The first is the analysis of ACTIVATE mission data to explore effects of aerosol layer height on photolysis rates. The second is utilizing PACE mission data as mock-PolCube data to create a photolysis rate product pipeline for future PolCube operations.

Methods

The methodology of retrieval of the aerosol layer heights vary between each of the instruments used in the study. Firstly, looking at the instruments on ACTIVATE and the difference between the RSP and HSRL-2 layer heights. HSRL-2 aerosol layer heights are flagged as "truth" heights for comparison with the RSP heights. To derive the aerosol layer height, the Haar wavelet covariance transform is used to find gradients in the aerosol backscatter profiles measured by HSRL-2[13, 14]. The viewing geometry of looking nadir (directly down) from a plane means that the retrieved layer height is the top of the aerosol layer. RSP takes advantage of its multiple viewing angles to measure the polarization for many scattering angles. Working better when looking down at a cloud layer with strong polarization features, RSP looks for subtle changes in the polarized spectra that get distorted through interaction with an aerosol layer. The lower the layer is, i.e., closer to the cloud layer, the more distortion, and opposite for high layers. In contrast to HSRL-2, which retrieves the top of the layer, an especially diffuse aerosol layer will cause RSP to retrieve a "mixed" layer height that may fall deeper within the layer boundaries. These are both different than the method used with PACE's polarimeters. PACE retrieves its aerosol layer height (this is one of many methods PACE uses) by measuring two strong oxygen absorption lines from light scattered off of an aerosol layer[12]. Since the amount of oxygen is known in the entire atmosphere, the amount of light absorbed away on the trip back to the instrument after scattering can be equated to the height of the aerosol layer. The lower the layer, the more oxygen absorption.

Fast-J requires aerosol microphysical properties at 27 different wavelengths from UV-IR to run. RSP measures in 9 fewer wavelength bands, and PACE measures fewer than that. To overcome this, the aerosol properties are extrapolated to the missing wavelengths prior to running Fast-J.

Figure 3 shows an example of RSP wavelength extrapolation to the longer wavelengths Fast-J needs. The AOD is assumed to follow a

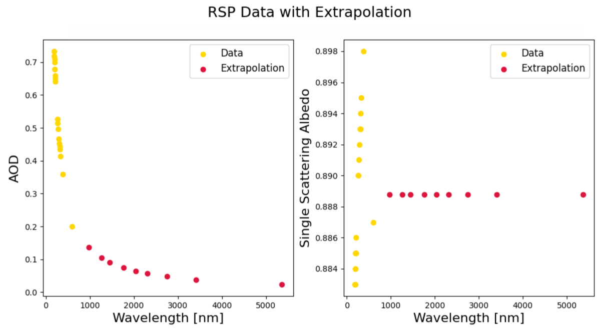

Figure 3: This figure shows the extrapolation of AOD (left) and SSA (right) for an example of RSP data from ACTIVATE.

power law, and the mean of the SSA is used for the extrapolated wavelengths. The lower energy radiation will have a harder time causing photochemical reactions, but it is still important to track how the extrapolation is done. PolCube will have a wider range of wavelengths than both RSP and PACE and will not need as much assumptions in the extrapolation of properties.

Once all 27 wavelengths have data to be put into Fast-J, the algorithm can then be run for analysis of height sensitivities as well as how the PolCube/PACE pipeline performs.

Results

For the ACTIVATE analysis, Figure 4 shows the percent difference of photolysis rates for the destruction of NO2 from urban pollutant aerosol layers gotten from HSRL-2 (truth) and RSP (test). The plot is also colored by the height difference between the co-located HSRL-2 and RSP measurements in kilometers. We see that there can be a large variation in surface photolysis rates (hundreds of percent) when the two height measurements from HSRL-2 and RSP don't meet. There is much less when the height measurements agree better, but there can still be up to a 100 percent difference when the heights differ by a few hundred meters to a kilometer.

Figures 5 and 6 show the results of the PACE photolysis rate retrieval through a map of surface photolysis rates and the profiles of each pixel seen on the map. Both figures showcase the results of running data from the local smoke event on June 4th 2025, when Canadian wildfire smoke traveled south over Hampton Roads.

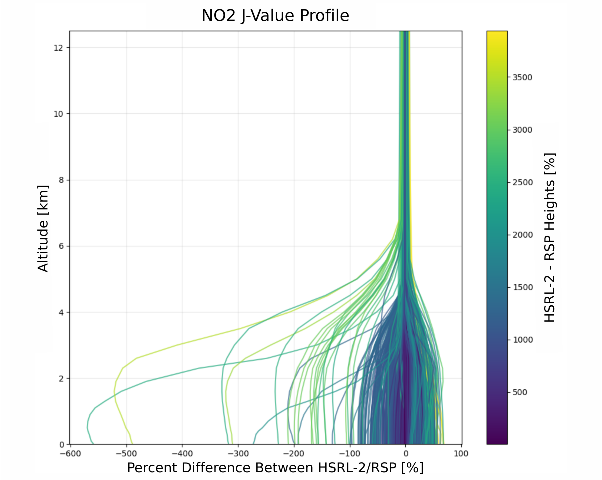

Figure 4: Percent differences between the HSRL-2-derived-height photolysis rates (NO2 destruction) and the RSP-derived-height photolysis rates as a function of height and colored by the separation between HSRL-2 and RSP heights in km.

Discussion

Figure 4 shows that choosing the instrument you use to retrieve aerosol layer height can influence the photolysis rates calculated using Fast-J. As discussed in the methods section, the two measurements HSRL-2 and RSP can differ due to the physical nature of the measurement. This is why it is helpful to have both instruments on board a flight campaign like ACTIVATE. In this case, the Lidar can be used as the ”true” height, while RSP can only be used for the microphysical aerosol properties. However, PACE (and soon PolCube) orbits without a lidar onboard. This means we must rely on the polarimeter to make the aerosol layer height measurement. To explore this in the future, I will develop and train a neural network with

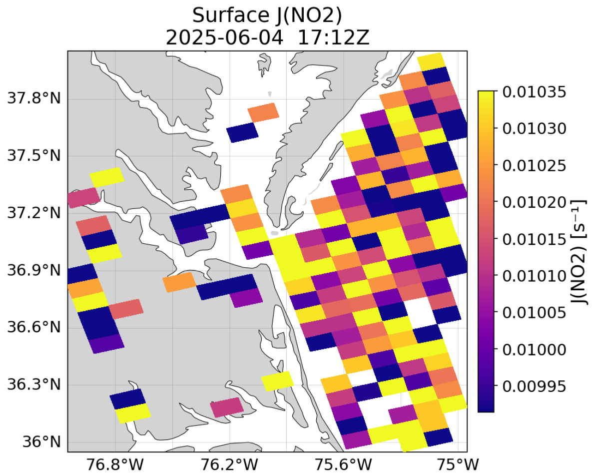

Figure 5: Map of Hampton Roads area with the surface photolysis rates (for NO2 destruction) from the 06/04/2025 smoke event.

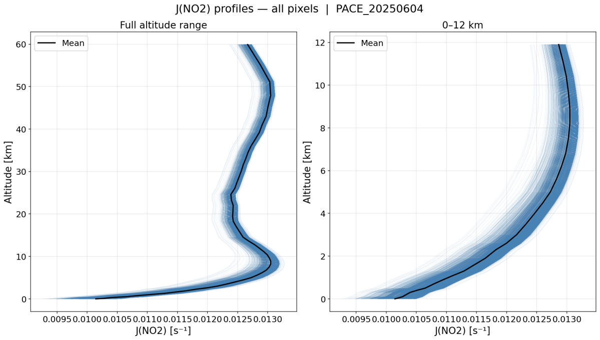

Figure 6: Photolysis rate profiles (NO2 destruction) for each pixel of the image from Figure 5, along with the mean value.

layer heights from HSRL-2 and test it with the heights from RSP with the goal of improving photolysis rate products from polarimeters. A network trained on lidar heights that gives polarimeters higher accuracy would allow accurate photolysis products coming from orbiting polarimeters that can span across an extensive spatial coverage.

The PACE photolysis rate pipeline shows a proof of concept that can be easily tweaked for use with the PolCube CubeSat. In the case of PACE, there are more methods that others have created for retrieving the aerosol layer heights. These other methods will also be analyzed as part of the pipeline. There are also studies underway that combine PACE data with flight campaigns to validate PACE retrievals that could help improve the accuracy of the pipeline.

Conclusion

Aerosols and their properties have large and quantifiable effects on the photolysis rates of many species in the atmosphere. Understanding how this alters the chemistry of the troposphere, specifically here in the boundary layer, is vital for public health.

The PACE photolysis rate pipeline is a strong proof of concept for a photolysis rate pipeline for PolCube and allow for more accurate photolysis rates to be calculated to then be put into global chem models such as GEOS-CHEM and WRF-CHEM.

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