

# Ice formation in the exhaust plume from an Ariane5 rocket

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## Abstract

Rockets directly inject water vapor and aerosol into the atmosphere, promoting ice nucleation and formation of cirrus clouds in cold, ice supersaturated regions of the atmosphere. Enhanced ice cloud occurrence frequency has been detected in the mesosphere near 80 km altitude after the launches of space shuttles and smaller rockets. Here, we present unique evidence for ice cloud formation at lower altitudes caused by ice nucleation in the exhaust plume from an Ariane5 launch vehicle. The rocket condensation trail has been detected in high resolution data products from the SEVIRI Spinning Enhanced Visible and Infrared Imager on the METEOSAT-9 satellite in a region south of the rocket launch site near Kourou, French Guiana. Meteorological reanalysis data from the European Centre for Medium-Range Weather Forecasts show significant ice supersaturation in that region in the tropical tropopause near the 100 hPa level. We suggest that ice nucleated predominantly on particulate aluminum oxide emissions in the rocket exhaust plume. We investigate the ice nucleation pathway and follow the evolution of the rocket contrail in the upper troposphere and lower stratosphere for almost two hours.

## 1. Introduction

Combustion of solid  $\text{NH}_4\text{ClO}_4/\text{Al}$  material in rocket propulsion systems leads to the emission of enhanced number densities of solid particles into the atmosphere, predominantly consisting of  $\text{Al}_2\text{O}_3$  material. In the atmosphere, the emission products in the hot and humid rocket exhaust plume mix with ambient air. Thereby the exhaust cools down and the relative humidity in the plume increases. Eventually, in cold regions of the atmosphere the plume reaches saturation conditions with respect to liquid water and the solid or processed coated plume particles may get activated into small water droplets. Ice nucleates in the activated water droplets as soon as the plume is ice supersaturated. The initially small ice particles grow by uptake of ambient water or by aggregation with other ice particles. In addition, the plume particle concentration dilutes due to entrainment of ambient air. If the ambient air is supersaturated with respect to ice, the ice particles survive and grow further. This process has been observed to promote polar mesospheric cloud formation after the launch of rockets. If the ambient air is sub-saturated with respect to ice, the rocket contrail particles may sublimate. The remaining solid  $\text{Al}_2\text{O}_3$  aerosol can be transported over long distances and may modify ice clouds during their life cycles until they are finally removed from the atmosphere by rain out or snow out processes.

In the following, we compile previous observations of aerosol particles in rocket plumes and of ice cloud formation in the mesosphere related to the launch of space vehicles. We show a unique satellite observation of ice cloud or contrail formation at altitudes below 30 km in the exhaust plume from an Ariane5 rocket. We investigate the ice formation process and follow the contrail evolution over few hours. Finally, we discuss implications of rocket contrail formation for atmospheric composition and climate.

## 2. Aerosol size and composition in rocket exhaust plumes

Exhaust plumes from several rocket engines including a space shuttle launch vehicle, a Titan IV, an Athena II, an Atlas II and a Delta II rocket have previously been probed with instruments on the NASA WB57 high altitude research aircraft. Particle number concentrations of several hundred per  $\text{cm}^3$  of  $\text{Al}_2\text{O}_3$  particles in the size range of 0.01 to 4  $\mu\text{m}$  have been observed in 5-min old stratospheric exhaust plumes from two solid rocket motor space shuttle launch vehicles and a Titan IV rocket [1]. The observations suggest a trimodal particle size distribution of the plume with mode mean diameters near 0.005  $\mu\text{m}$ , 0.1  $\mu\text{m}$ , and 2  $\mu\text{m}$ . Similar aerosol concentrations of few hundred per  $\text{cm}^3$  have been measured in the stratospheric exhaust plume from a Delta II rocket powered by a combination of solid  $\text{NH}_4\text{ClO}_4/\text{Al}$  and liquid  $\text{LOx}/\text{kerosene}$  propulsion system [2]. In the plume the injection of chlorine species as combustion product from the  $\text{NH}_4\text{ClO}_4/\text{Al}$  fuel and their activation by sunlight promotes ozone depletion in the plume. Complete ozone loss probably initiated by gas phase reactions of the catalytic  $\text{ClO}$  dimer cycles at warm temperatures or by heterogeneous activation on solid or liquid plume particles has been observed in the Delta II rocket plume.

Particle number, size and composition measurements have been performed in the stratospheric exhaust plume of a small Athena II SRM at less than 30 min plume age [3]. The plume contained alumina particles with number densities near 10000 per  $\text{cm}^3$  in the size range between 0.004 and 1.2  $\mu\text{m}$ . More than 2300 exhaust particles from the Athena II SRM plume and the plume from the space shuttle launch vehicle have been probed and analyzed with an aerosol composition mass spectrometer [4]. The rocket plume aerosol contained primary and trace components of the aluminum fuel and the combustion catalyst. In addition, chlorine from the oxidizer, iron and elemental carbon were found in the Athena II rocket plume and nitrate, phosphate and water in the space shuttle plume particles. These observations suggest that the particle composition in the exhaust plumes is very diverse and that solid plume particles may get activated or coated with condensable exhaust products. The detection of components of the rocket fuel, oxidizer, binding agents, and the combustion catalyst in the aerosol particles or on particle surfaces leaves open question regarding heterogeneous chlorine chemistry on coated alumina particles and their reactivity.

Enhanced concentrations of reactive nitrogen species ( $\text{NO}_y$ ) have also been observed in the Athena II SRM plume [5]. High nitric acid mixing ratios indicate that  $\text{HNO}_3$  is a significant plume component, most probably formed by the heterogeneous reaction of  $\text{ClONO}_2$  and  $\text{HCl}$  on the emitted alumina particles. High nitric acid concentrations at temperatures below 215 K may favour the formation of solid nitric acid trihydrate (NAT) particles in rocket exhaust plumes [6] which has frequently been observed in the polar stratosphere [7]. A similar NAT cloud formation process may occur in stratospheric rocket wakes.

The injection of chlorine species as combustion product from the  $\text{NH}_4\text{ClO}_4/\text{Al}$  fuel and their activation by sunlight may promote ozone depletion in the plume. Complete ozone loss probably initiated by gas phase reactions of the catalytic  $\text{ClO}$  dimer cycles at warm temperatures has been observed to occur within 1 hour [2]. In addition, heterogeneous reactions on processed  $\text{Al}_2\text{O}_3$  particles, as well as on solid NAT or ice in the exhaust plume particles may activate chlorine and enhance ozone loss.

The particle emissions from rocket exhaust plumes in the stratosphere can be compared to the meteoritic influx as natural source of particles into the stratosphere. Most of the meteoritic mass influx in the atmosphere of 8 to 30 Gg per year ablates at altitudes above 80 km due to frictional heating [8 and references therein]. The ablation products recondense, coagulate in the mesosphere and form nanometer-sized smoke particles. These meteoritic smoke particles are transported with the atmospheric circulation and sediment down. They remain about a year in the stratosphere, become well-mixed and are partly incorporated into stratospheric sulfate aerosol.

Compared to the meteoritic influx, the total rocket propellant emission from a fleet of space shuttle, Titan IV, Proton, Long March, Ariane5, Soyuz, Zenit and smaller rocket launch vehicles has been calculated to amount to about 30 Gg per year averaged over 2000 to 2002 of which about 9 Gg per year are emitted into the stratosphere [9]. The particulate alumina fraction of the rocket emissions in stratosphere is about one third of the total emissions and amounts to 3 Gg annually, hence the solid particle rocket influx is significantly smaller than meteoritic influx.

### 3. Polar mesospheric cloud formation induced by rocket emissions

Enhanced polar mesospheric cloud (PMC) formation has been detected after the launch of several small rockets and large space shuttle launch vehicles. An increase in the mesospheric water concentration caused by rocket emissions has been measured with the Sounding of the Atmosphere with Broadband Emission Radiometry (SABER) instrument on the Thermosphere Ionosphere Mesosphere Energetics and Dynamics (TIMED) satellite [10]. In 2002 the radiometer detected radiance enhancements in altitudes of 90 and 130 km related in time and location to emissions from space shuttles or from liquid fuelled rockets. Almost half of the total liquid fueled launches, including all space shuttle launches were identified in the radiance data and lead to enhanced water vapor layers in the mesosphere.

An Arctic mesospheric OH layer has been observed a day after a space shuttle launch in 1997 at altitudes above 85 km. At these altitudes OH is produced predominantly by the photolysis of water vapor, hence it can be used as indicator for water vapor [11]. Exhaust from the shuttle's main engines is almost entirely water vapor and about 300 Gg of water vapor are injected into the altitude region between 108 and 114 km. This corresponds to water vapor mixing ratios of 3.5 ppmv in that altitude range. One week later a polar mesospheric cloud has been detected at latitudes between 70 and 75°N with the CRYogenic Infrared Spectrometers and Telescopes for the Atmosphere-Shuttle PAllet Satellite (CRISTA-SPAS) instrument on the Discovery payload [11]. Trajectory calculations link the PMC observation site to the initial water vapor enhancement by the space shuttle exhaust. Hence the observations suggest that rocket exhaust plumes can significantly perturb the mesospheric water vapor and cloud budget.

Transport of water vapor from a space shuttle exhaust plume to the southern hemisphere has been observed by the Global Ultraviolet Imager on the TIMED satellite [12]. The instrument detects mesospheric water vapor using scattered solar Lyman  $\alpha$  radiation. Mesospheric water vapor emissions from the Columbia space shuttle launched from Kennedy Space Center in January 2003 were followed over 2 days, initially spreading and moving north-east. After one day they change to southern directions reaching 35°S. Ground-based light detection and ranging (lidar) observations from Rothera, Antarctica, show iron layers near 112 km altitude 3 to 4 days after the space shuttle launch with high iron concentrations reaching  $1.5 \cdot 10^4 \text{ cm}^{-3}$ . As there is no known natural source of neutral iron above 100 km, these observations have been interpreted as iron ablated from the shuttle's main engines.

Lidar, radar, and optical observations have also been performed in Alaska in the polar summer mesosphere shortly after a space shuttle launch in 2007 [13]. An exceptionally intense mesospheric cloud has been detected 3 days after the launch of the space shuttle on 8 August. Similar to previous observations, the space shuttle launch was followed by enhanced polar mesospheric cloud formation and the build-up of an iron layer. The observed iron layer increased by a factor of 20 in density compared to background levels. Co-located with the iron layer, the radar records detected polar mesospheric summer echoes in this event.

### 4. Contrail formation in an Ariane5 rocket exhaust plume

While formation of ice particles related to rocket exhaust plumes has been reported previously in the mesosphere, we present first evidence for ice nucleation in rocket plumes in the tropopause region near and below 20 km altitude. When the hot aircraft exhaust mixes with ambient cold air and it cools down. When saturation with respect to water is reached, water condenses on predominantly on solid  $\text{Al}_2\text{O}_3$  plume particles and forms small water droplets. When temperatures are below the ice frost point, ice nucleates in the activated aerosol. During further life time, the ice particles grow as long as the ambient air is supersaturated with respect to ice. The rocket contrails may survive for several hours and become persistent. The particle number density decreases due to inmixing of ambient air and the rocket contrail ice clouds lose their structure and may modify natural cirrus cloudiness. Here we present the unique case of contrail formation in the exhaust plume from an Ariane5 rocket. The Ariane5 has been launched on 26 November 2010 at 18:39 UTC at the Space Centre near Kourou in French Guiana.

The satellite analysis of this launch has been performed with the SEVIRI Spinning Enhanced Visible and Infrared Imager on board Meteosat-9 satellite positioned in geostationary orbit at 0°. The SEVIRI images provide information on solar and terrestrial radiation in 12 spectral channels. 11 of the spectral channels have a resolution of 3 km at the sub-satellite point while the High Resolution Visible (HRV) channel 12 provides data with a resolution of 1 km. In addition, the SEVIRI instrument has a high temporal resolution of 15 minutes, which is necessary for the launch

analysis and to investigate the cloud life cycle. As clouds and rocket plumes are small scale and fast moving objects, we employ the high temporal and spatial resolution provided by the HRV channel. As the HRV channel is located in the visual spectrum, no information is available during night time.

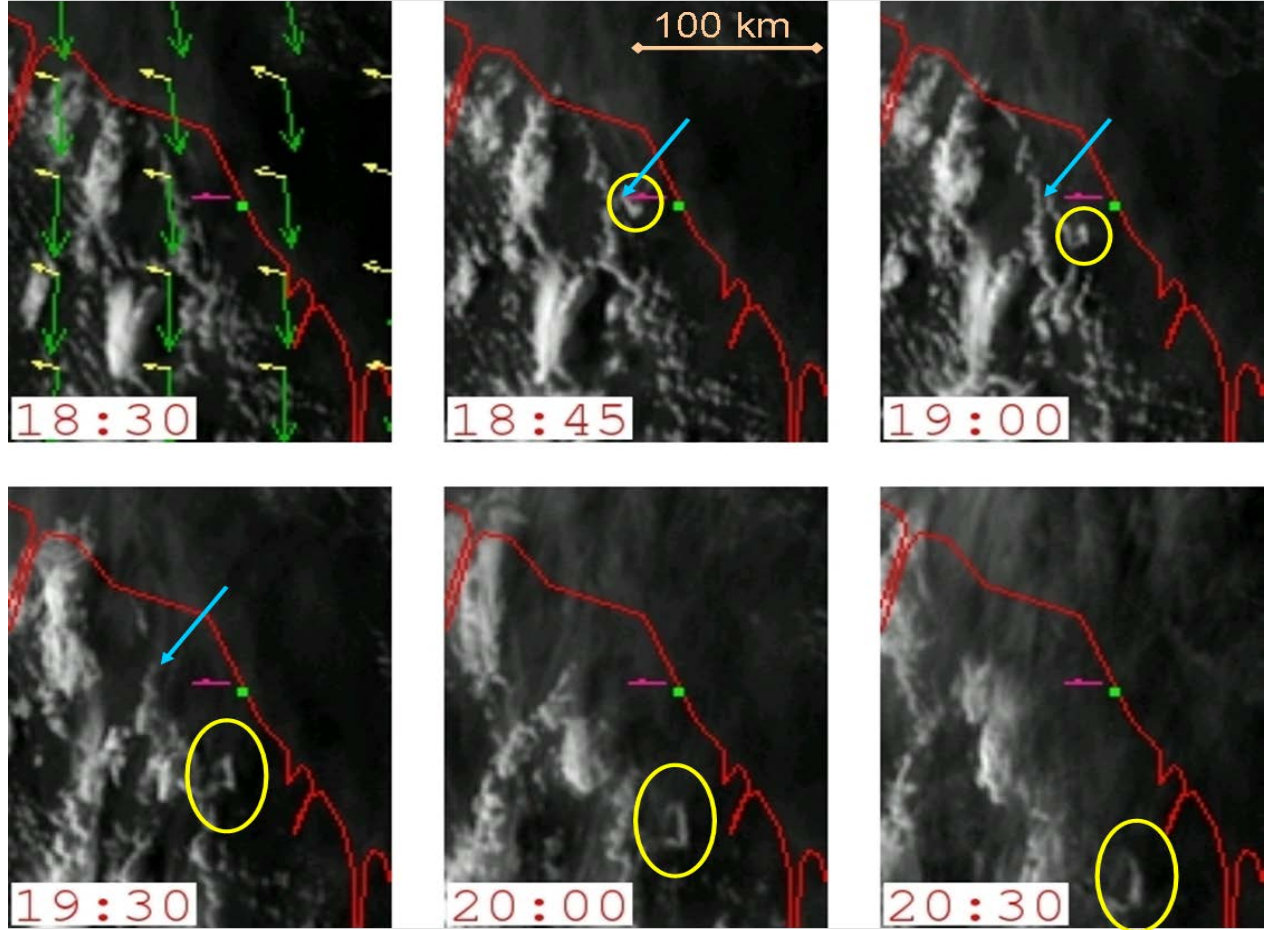


Figure 1: Satellite images from METEOSAT-9 SEVIRI instrument, HRV channel on 26 November 2010.

All scenes: Green dots indicate position of Kourou City in French Guyana.

The pink line represents trajectory of the Ariane5 launch vehicle from 0 to 20 km altitude.

Whereas Ariane5 originally started eastwards, trajectory is seen westwards due to parallax effects.

Scene at 18:30 UTC (before start):

Mainly natural cloudiness, wind direction in 850 hPa (yellow arrows), wind direction in 100 hPa (green arrows).

Scenes at 18:45 UTC and thereafter:

The yellow circles indicate position of the Ariane5 rocket contrail near 14 km altitude.

The blue arrows mark the position of the low-level rocket plume below 4 km altitude.

Figure 1 shows 6 high resolution visible images at 15 or 30 minute time steps between 18:30 and 20:30 UTC on 26 November 2010. At 18:30 UTC, 9 minutes before the rocket launch, mainly natural cloudiness is detected by the satellite instrument. In 850 hPa near the ground the wind direction is to the west (yellow arrows) and in 100 hPa the wind direction is to the south (green arrows). The location of the launch site Kourou is marked by a green dot. The Ariane5 flight track from the surface to 20 km altitude is marked by a pink line in each image.

After the Ariane5 launch at 18:39 UTC, two effects of rocket emissions can be observed in the satellite images. At 18:45 UTC a line-shaped cloud can be identified and related to the launch site and the rocket emissions. The line-shaped structure marked by the yellow circles drifts in south-eastern direction. The rocket contrail is visible in each image within the next 2 hours. In addition, there is an exhaust plume marked with the blue arrow drifting westwards with the wind direction at lower altitudes.

To obtain information on the altitudes of these two objects, we use reanalysis data from numerical weather prediction model operated by the European Centre for medium-range weather forecast ECMWF, which integrates information from different observational sources into the data assimilation. The ECMWF wind fields at low altitudes are directed towards the west, hence support the visual impression that the westward moving cloud structure (blue arrows) is caused by the particulate rocket exhaust plume near take off in altitudes below 2 to 4 km.

In contrast, the line-shaped contrail structure drifts in southern direction. ECMWF reanalysis of wind fields show winds from north only in a small altitude range represented by the 100 hPa pressure level. This corresponds to an altitude of 14 km hence above the level of local air traffic.

In addition, we investigate the relative humidity fields in the upper troposphere and tropopause region on that day in order to evaluate the potential of ice cloud and contrail formation. The relative humidity data are again provided by ECMWF reanalysis. Figure 2 shows the relative humidity with respect to water above 100% in a large area above French Guiana, which strongly supports the possibility of rocket contrail formation. Within the next 2 hours the rocket contrail can be identified in each satellite image. It drifts in southern directions till darkness prevents the detection of its further evolution.

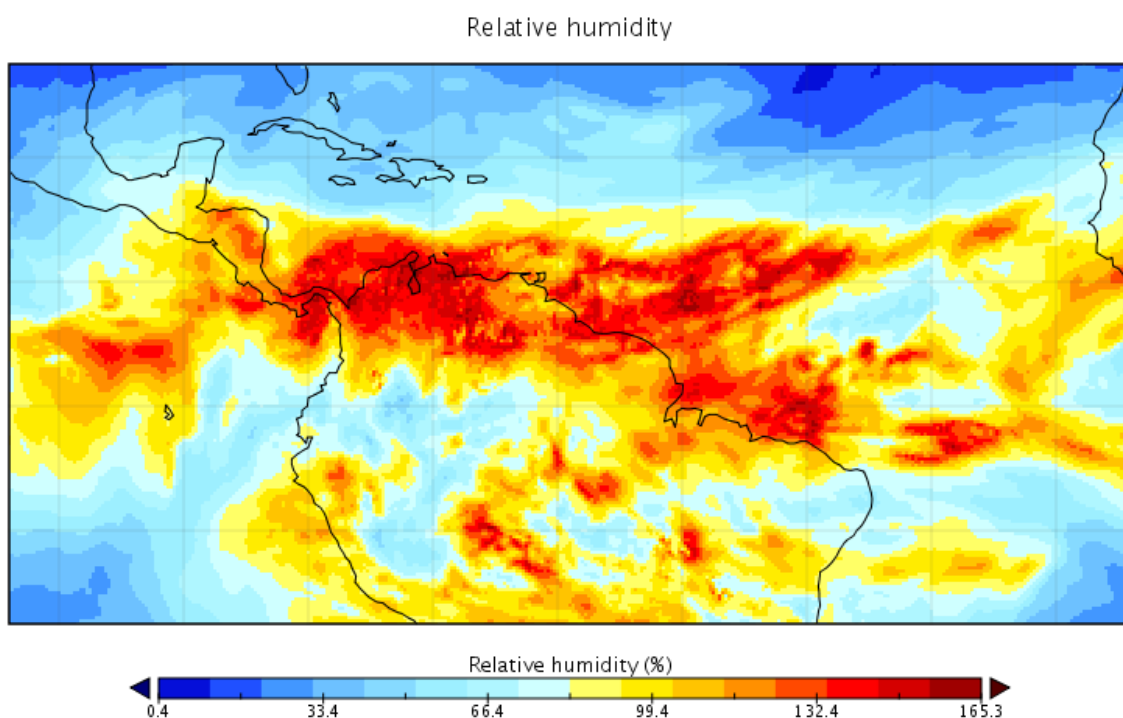


Figure 2: Relative humidity on 26 November 2010 at 18:00 UTC from ECMWF reanalysis at the 100 hPa level

## 6. Summary and outlook

Strong local and regional perturbations of atmospheric trace gas and particle fields have been detected in recent years after the launch of rockets by in-situ instruments on aircraft and by remote sensing observations from satellite. Enhanced water vapor and iron levels have been observed after the launch of space shuttles and large rockets followed by enhanced mesospheric cloud formation both in polar and extra-polar regions of the mesosphere. Emission of alumina particles and chlorine compounds from SRM can cause complete ozone destruction in the young exhaust plume and potentially lead to a global total ozone loss  $< 1\%$ . Still insufficient knowledge on ozone loss processes on the processed plume particles and on rocket contrails renders a final conclusion on global ozone loss unknown.

Here, we present the first direct evidence for ice formation in the upper troposphere caused by rocket emissions. We suggest that ice nucleation on solid or processed  $\text{Al}_2\text{O}_3$  particle emissions is the dominant ice formation pathway in

rocket plumes. New in-situ data on humidity, aerosol and ice particle size distributions in rocket contrails in combination with process modeling would be required to investigate the ice nucleation process in rocket contrails in more detail. The unique observation of an Ariane5 rocket contrail in 14 km altitude and the detection of a particulate rocket aerosol layer at ground levels from Meteosat SEVIRI images should be verified in other rocket launch scenarios in order to better quantify the local and global effects of rocket emissions on the atmosphere. Due to the local cloud structure and caused by restricted rocket launch rates, we estimate that the global impact of rocket contrails on atmospheric composition and climate is very small. Still, the combined evidence from the satellite images and ECMWF reanalysis data suggests that contrails can form on rocket exhaust plume particles and that rocket contrail formation can be detected in meteorological geostationary satellite data.

## References

- [1] Ross, M. N., P. D. Whitefield, D. E. Hagen, and A. R. Hopkins. 1999. In situ measurement of the aerosol size distribution in stratospheric solid rocket motor exhaust plumes. *Geophys. Res. Lett.* 26:7–13 doi:10.1029/1999GL900085.
- [2] Ross, M. N., et al. 1999. Observation of stratospheric ozone depletion associated with Delta II rocket emissions *Geophys. Res. Lett.* 27:15–19. doi:10.1029/1999GL011159.
- [3] Schmid, O., J. M. Reeves, J. C. Wilson, C. Wiedinmyer, C. A. Brock, D. W. Toohey, L. M. Avallone, A. M. Gates, and M. N. Ross. 2003. Size-resolved particle emission indices in the stratospheric plume of an Athena II rocket. *J. Geophys. Res.* 108:4250–4256. doi:10.1029/2002JD002486.
- [4] Cziczo, D. J., D. M. Murphy, D. S. Thomson, and M. N. Ross. 2002. Composition of individual particles in the wakes of an Athena II rocket and the space shuttle. *Geophys. Res. Lett.* 29:2047–2051. doi:10.1029/2002GL015991.
- [5] Popp, P. J., et al. 2002. The emission and chemistry of reactive nitrogen species in the plume of an Athena II solid-fuel rocket motor. *Geophys. Res. Lett.* 29:1887–1889. doi:10.1029/2002GL015197
- [6] Gates, A. M., et al. 2002. In situ measurements of carbon dioxide. 0.37–4.0  $\mu\text{m}$  particles, and water vapor in the stratospheric plumes of small rockets. *J. Geophys. Res.* 107:4649–4656. doi:10.1029/2002JD002121.
- [7] Voigt, C., J. Schreiner, A. Kohlmann, P. Zink, K. Mauersberger, N. Larsen, T. Deshler, C. Kröger, J. Rosen, A. Adriani, F. Cairo, G. Di Donfrancesco, M. Viterbini, J. Ovarlez, H. Ovarlez, C. David, A. Dörnbrack. 2002. Nitric Acid Trihydrate (NAT) in Polar Stratospheric Clouds. *Science*. 290:1756–1758.
- [8] Cziczo, D. J., D. S. Thomson, and D. M. Murphy. 2001. Ablation, flux and atmospheric implications inferred from stratospheric aerosol. *Science* 291:1772–75.
- [9] Jackman, C., D. Considine, and E. Fleming. 1996. Space shuttles impact on the stratosphere: An update. *J. Geophys. Res.* 101(D7). doi:10.1029/96JD00577.
- [10] Siskind, D. E., M. H. Stevens, J. T. Emmert, D. P. Drob, A. J. Kochenash, J. M. Russell, L. L. Gordley, and M. G. Mlynczak. 2003. Signatures of shuttle and rocket exhaust plumes in TIMED/SABER radiance data. *Geophys. Res. Lett.* 30:1819–1823. doi:10.1029/2003GL017627.
- [11] Stevens, M. H., J. Gumbel, C. R. Englert, K. U. Grossmann, M. Rapp, and P. Hartogh. 2003. Polar mesospheric clouds formed from space shuttle exhaust. *Geophys. Res. Lett.* 30:1546–1550. doi:10.1029/2003GL017249.
- [12] Stevens, M. H., R. R. Meier, X. Chu, M. T. DeLand, and J. M. C. Plane. 2005. Antarctic mesospheric clouds formed from space shuttle exhaust. *Geophys. Res. Lett.* 32. L13810. doi:10.1029/2005GL023054.
- [13] Kelley, M. C., M. J. Nicolls, R. H. Varney, R. L. Collins, R. Doe, J. M. C. Plane, J. Thayer, M. Taylor, B. Thuraijah, and K. Mizutani. 2010. Radar, lidar, and optical observations in the polar summer mesosphere shortly after a space shuttle Launch. *J. Geophys. Res.* 115. A05304. doi:10.1029/2009JA014938.