# Current Environmental Concerns about Space and Suborbital Launch Activities*

Filippo MAGGI,[1)†] and Stefania CARLOTTI[1)]

[1)]*Dept. Aerospace Science and Technology, Politecnico di Milano, Milano, 20156, Italy*

The development of the Space economy and the consequent increment of launch service request raised the concern about the environmental sustainability of the sector. Despite a more realistic impact evaluation on climate change would require a wider perspective (typical of Life Cycle Analyses), the scientific community mainly lends emphasis to rocket atmospheric emissions. This interest developed systematically only after the advent of Space Shuttle and progressively grew since the beginning of this century. Initially, the focus was mainly posed on chorine-based exhaust products and on their impact on ozone layer. Nowadays it is clear that the picture is more complex and includes particle emission, soot, radiative forcing contribution, and ozone depletion by catalytic effects. This paper is an initial review about the current knowledge on rocket atmospheric emissions and, without the aim of being universal, suggests some areas where deeper comprehension should be developed.

**Key Words:** Rocket propulsion, environmental impact, plume

## Nomenclature

| | | |
|---|---|---|
| $h$ | : | altitude |
| $AP$ | : | ammonium perchlorate |
| $B$ | : | booster |
| $GHG$ | : | greenhouse gas |
| $HTPB$ | : | hydroxyl-terminated polybutadiene |
| $NH$ | : | northern hemisphere |
| $OF$ | : | oxidizer-to-fuel mass ratio |
| $RP1$ | : | rocket propellant 1 |
| $S1$ | : | first/core stage |
| $S2$ | : | second stage |
| $SSME$ | : | Space Shuttle Main Engine |
| $UDMH$ | : | unsymmetrical dimethyl hydrazine |

## 1. Introduction

Literature about the environmental impact of space propulsion launchers is quite sparse and research efforts were not constant in time. It appears that most of the interest was raised once the Space Shuttle program came to service. Most of the concern was focused on stratospheric depletion of ozone above the Earth pole regions. At that time only few research activities were related to emissions into the lower atmosphere, toxicity, ground contamination and similar. Most of the works focused on initial assessments of the environmental impact for the Space Shuttle in its early flight phase and in the surrounding areas.[1,2)] On this latter aspect, military literature covered the topic better than the civilian one, discussing both gun and rocket propellant emissions connected to toxicity issues for battlefield operations.[3)]

The interest on atmospheric effects by rocket emissions grew progressively in time but it was never considered a priority by the scientific community. As a consequence of the "Montreal Protocol on Substances that Deplete the Ozone Layer (1987)" attention was posed on released chlorine, first. In the 1991 Scientific Assessment of Ozone Depletion, an entire chapter was dedicated to Space Shuttle and rockets in general, testifying the increased attention.[4)] In the same report, some discussion was spent also for the emitted particulate but the authors underlined the lack of specific knowledge. Few years later, a report by the Aerospace Corporation suggested three possible influencing processes between particulate and ozone depletion in presence of chlorine.[5)] The interaction was later confirmed by laboratory experiments.[6)] In the last 20 years the knowledge of atmospheric pollutant evolution grew consistently. Both aluminum oxide and carbon black particles emitted by rockets and aircrafts became the subject of studies about climate change and ozone depletion. For example, Ross and co-authors developed climatological studies considering increasing rocket launches due to expansive scenarios of the space economy and warned about the possible consequences of multiple daily launches from same locations using propulsion systems with high carbon black emission index.[7)]

To date, the current launch rate does not represent yet a threat, if compared with other anthropogenic sources of pollution. However, market predictions forecast a doubling/tripling of market size by 2027, meaning a consistent increment in the number of launches. At the same time, rocket emission data and climatological predictions lack of precise characterization and full understanding. In this respect, a very interesting and updated discussion is reported in a public document released by The Aerospace Company and authored by Ross and Vedda.[8)]

Without the aim of a complete review, the present paper explores the current knowledge of pollution emission, looking at existing experimental and modeling data about the emissions of thermochemical propulsion units. The discussion refers to solid, liquid, and chemical-hybrid propulsion systems. This work grounds on four recent documents (Ross and Vedda,[8)] Dallas and co-authors,[9)] Murray and co-authors[10)] and Voigt and co-authors[11)]) which supply an updated and general vision of the pollution generated by space launch activities.


†Corresponding author, filippo.maggi@polimi.it

## 2. Regions of pollution

The emissions of rocket launchers can be classified in different manners. At first, we can consider the region of the atmosphere in which they are released.

Emissions in troposphere The troposphere extends from ground to about 10-18 km, depending on the latitude. In this part, pollutants are released in proximity of the launch site and in the lower part of the atmosphere. Their effect is reportedly local and the residence time of these pollutants is small since rain and fall-out make them fall back to the earth. Moreover the dynamics of the troposphere is characterized by global circulations vortices and a negative temperature gradient which favor short air turnover and condensation of gases characterized by relatively high boiling points (e.g. water vapor). In addition, ground contamination from first stage reentry and spillage of propellant leftovers have been signaled[12] but this specific aspect is beyond the scope of this paper. In this region of the atmosphere, launcher architectures opt for the use of booster stages and main stages. We can find examples of any type of chemical propulsion systems (solid, liquid cryogenic, liquid storable hypergolic or non-hypergolic, and potentially chemical-hybrid).

Stratospheric emissions The stratosphere extends up to about 50 km of altitude and is dynamically isolated from the troposphere, making it more sensitive to direct emissions.[8] The dynamics of the stratosphere is ruled by a reverse temperature gradient, favoring stratification. A global circulation process involving also the troposphere (Brewer-Dobson) exists between the equatorial region and the poles.[13] The overturn of air in this region changes with the altitude and was recently estimated by Linz and co-authors to be around 6 months to 1.5 years.[14] In an earlier work, Waugh and Hall reported a much larger time frame, estimating up to about 5 years, with higher latitudes characterized by much longer turnover.[15] In general, in this part of the atmosphere initial stages complete their missions, and upper stages ignite.

Mesospheric emissions The mesosphere extends above the stratosphere up to about 85-100 km, where another temperature minimum is reached. Under such frosty conditions mesospheric clouds can be generated by water vapor in proximity of the poles. The interest over mesosphere rocket emissions is quite recent and followed the publication of works connecting the generation of said clouds to rocket launch activities. For example, Stevens and co-authors could trace the origin of a mesospheric cloud to the launch of Space Shuttle STS-85.[16]

## 3. Type of pollutants

Typically, space launchers are characterized by multistage architectures, consisting in either parallel or tandem configurations. Table 1 reports a limited selection of launch vehicles and of respective stages used below the Karman line. Stages operating above the mesosphere are neglected. We can identify three main rocket propulsion categories. Solid propellants are mostly used for boost stage of heavy launchers (Shuttle and Ariane V) and, in the case of VEGA launcher, also for upper stages. Cryogenic propulsion units based on hydrogen and oxygen operate across the entire trajectory and, for the selected examples, work up to orbital condition. RP1/oxygen units also operate across the entire mission. The Spaceship 2, a space plane operated by Virgin Galactic for suborbital touristic flights, is air-launched by a subsonic aircraft and is powered by a hybrid propulsion unit using nitrous oxide oxidizer and a polymer fuel. Its propulsion mission is the shortest as the engine powers a suborbital mission.

Table 1. Examples of stage operational altitude in troposphere,stratosphere and mesosphere

| Launcher | Stage | h, km |
|---|---|---|
| VEGA (VV-10) | S1 - SRM | 0 - 59 |
| | S2 - SRM | 59 - 155 |
| Ariane V ECA(GTO) | B - SRM | 0 - 69 |
| | S1 - LOX/LH2 | 0 - 178 |
| Falcon 9 (CRS-3) | S1 - LOX/RP1 | 0 - 80 |
| | S2 - LOX/RP1 | 80 - orbit |
| Space Shuttle (STS-30) | B - SRM | 0 - 47 |
| | SSME - LOX/LH2 | 0 - orbit |
| Spaceship 2 (Unity 22) | Carrier | 0 - 14 |
| | S1 - $N_2O$/Polymer | 14 - 40 |

Rocket pollutants derive either from direct combustion or from plume interaction with the atmosphere (afterburning). Thermodynamics supplies only an approximate estimation of the exhaust gas composition as finite rate chemistry effects are not considered (see Table 2). For solid propellants based on ammonium perchlorate nozzle exhaust is made by water vapor, carbon dioxide and monoxide, hydrogen chloride, molecular hydrogen and nitrogen as gaseous main constituents. In addition, if aluminized propellants are used, aluminum oxide in solid form is produced. Hydrogen-fueled liquid propulsion units typically operate under fuel-rich conditions for optimization of the specific impulse, expelling water vapor and unburnt hydrogen. Hydrocarbon-based liquid propulsion units produce carbon monoxide and dioxide, water, and hydrogen. If a nitrogen-based propellant component is used (e.g. nitrous oxide or hydrazine) nitrogen is released as well. It is important to underline that combustion processes are not ideal and finite rate chemistry as well as flame structure may favor the generation of partial combustion products or soot. In addition, the interaction between plume and surrounding air (afterburning), the presence of supersonic dynamic patterns in the flow, and the reaction between gaseous species and solid particles can increment the local temperature and/or can favor the generation of reactive chemical species, such as oxides of nitrogen or nitric acid.[11,17]

All the propulsion systems do not have constant mass flow rate. Most of the liquid systems for core stages are throttleable and change the thrust level depending on mission constraints. A typical example was represented by the SSME system which performed a throttling maneuver across the max-Q flight condition, coming back to full thrust afterwards. Also chemical hybrid propulsion units can be engineered to perform throttleable maneuvers but, so far, such operating flexibility has been adopted in programs for the experimentation of extraterrestrial landers.[19,20] Solid propulsion boosters

Table 2. Nozzle exhaust predictions by thermochemistry.[18] Chamber pressure 7 MPa, expansion ratio 40,

| Oxidizer | LOX | LOX | N2O | AP |
|---|---|---|---|---|
| Fuel | RP1 | LH2 | HTPB | HTPB+Al |
| OF | 2.4 | 6.0 | 8.0 | 68/14/18 |
| CO | 33.2 | - | 2.5 | 25.8 |
| CO2 | 40.3 | - | 20.5 | 2.9 |
| H2O | 25.0 | 96.5 | 16.5 | 5.2 |
| H2 | 1.3 | 3.5 | 0.9 | 2.7 |
| HCl | - | - | - | 21.1 |
| N2 | - | - | 59.5 | 8.2 |
| Al2O3 | - | - | - | 34.0 |

or main stages do not have throttling capability (commanded thrust variation) but they feature pre-defined thrust modulation thanks to proper design of the propellant grain, making the discharge mass flow rate variable in time. Brady and co-authors reported that a generic flight of the Space Shuttle released about 35% of its propellant (ant its exhaust products) in the stratosphere.[5] Similar fraction was reported also for Ariane V in the range 15-60 km altitude. This rough quantification was based on trajectory simulations, considering that the booster separation occurs above 60 km, at the lower edge of the mesosphere. These figures of merit are quite different from the ones proposed by Ross and Sheaffer.[21] Their paper reports the shares of propellant consumed at different flight levels and a rough sum of the data results in 30% of propellant consumed below 15 km, 50% burned between 15 and 60 km and about 20% up to 100 km. It is likely that the authors neglected the propellant consumed above the Karman line.

## 4. Interaction with the atmosphere

The knowledge about the exhaust plume characteristics and the mechanisms of interaction with the atmosphere have grown consistently in the last 20 years. However, the competent literature still states that uncertainties are still too high for accurate evolution studies. Currently, there are two areas of main concern: radiative forcing and ozone depletion. Contribution to greenhouse gases is not part of the list.

Radiative forcing consists of the alteration of the incoming radiative flux to the Earth from the space. Most active elements are represented by particles (soot and alumina) which settle in the stratosphere for long time (months to years depending on size, density, and release location). Ross and co-authors presented a prediction where a global cooling effect on ground could be achieved by repeated launches of suborbital rocket mission characterized by high soot emission index.[7] The effect can be attributed to the generation of a layer of fine particles absorbing and radiating back to space part of the incoming radiation. The principle is also reported in geo-engineering studies which also suggest a potential negative effect on ozone.[22]

Ozone depletion mechanisms are based on free radical reactions involving species containing hydrogen (the so-called $OH_x$ reaction), nitrogen (the $NO_x$ reaction), and chlorine (the $ClO_x$ reaction) and having an odd number of electrons.[23] Rocket emissions can play a role in the production of reservoir molecules for chlorine (such as $HCl$) in the stratosphere or in the generation of hydrogen carriers ($CH_4$ or $H_2$) and nitrogen carriers (such as $N_2O$) both in troposphere and in stratosphere. In addition, solid particles suspended in the stratosphere were identified as potential catalysts for ozone depletion. A PhD thesis by Spencer measured the activity of $HCl$ reserve molecule on different substrates and observed a non-negligible activity on alpha-alumina.[24] Sullivan and co-authors studied the favorable decomposition of ozone in presence of alumina.[6] Also the soot emitted by hydrocarbon-based rockets can have role in ozone depletion. A model study by Bekki identified a potential mechanisms of catalytic heterogeneous effect between ozone and carbon black emitted by aircraft fleets in the upper troposphere, within certain borders of uncertainty.[25] According to the author, the model could justify the reduction of ozone in the northern hemisphere (NH). The author also noted the typical fractal shape of carbon black, leading to an increment of the specific surface area and representing one of the major uncertainty sources. In a very recent conference abstract, Maloney and co-authors identified another indirect mechanism of ozone reduction by carbon black. According to the public abstract, stratified soot provokes a change in temperature gradients and a modification in stratosphere dynamics, leading to global ozone loss. The authors claim that the work can document how hydrocarbon-based rocket engines can lead to ozone losses comparable to the ones produced by solid propulsion units emitting chlorine compounds.[26] It should be noted that a paper was not available at author's hands.

Most of the GHGs concentrate in the troposphere. Recent studies have highlighted a general cooling and thickening of the stratosphere as a consequence of troposphere warming[27] but the role of rockets is negligible. As a term of comparison, the reader should consider that the amount of propellant consumed (and emitted GHGs) by launchers is less than 1/10000 with respect to the fuel used by civil aviation. As a consequence, the radiative forcing of global greenhouse gases is reportedly several orders of magnitude larger with respect to the one generated by rocket emissions.[8]

About the contamination of the mesosphere, there are not so many data available and the knowledge is currently rather limited. The correlation between Space Shuttle launches and some polar mesospheric clouds derives from empirical observations supported by estimations of water vapor.[16] The generation of these clouds over the poles can occur even without nucleation sites and leads to a dehydration of the surrounding region. As a consequence, an increment of the night-time mesospheric ozone and a reduction of atomic hydrogen concentration were observed. The propagation of $H$ perturbation was seen to reach even the lower edges of the thermosphere but, according to Siskind and co-authors, this fact should not lead to consequences to the global hydrogen balance of the atmosphere.[28]

## 5. Importance of emissions

Only some chemical species create an interaction between plume and atmosphere. main ones are solid particles (carbon black and aluminum oxide emissions), chlorine, hydrogen, and nitrogen carriers. In this section an overview of some literature sources is performed on substances emitted by different propulsion technologies.

### 5.1. Aluminized solid propellant

Commercial solid propulsion units mainly use ammonium perchlorate as oxidizer, metal fuel and a rubber. Solid propellant emissions were largely analyzed in the history, even with in-situ plume analysis using high-altitude aircrfts. A consistent knowledge base was accumulated in time and identified chlorine compounds, aluminum oxide particles, and nitric acid as main pollutants.

Chlorine-based compounds The emission of chlorine compounds is a matter of propellant chemical composition. The decomposition of ammonium perchlorare leads to $HCl$ in gaseous form and, after the exhaust, most of the hydrogen chloride becomes hydrochloric acid $HCl(H_2O)_x$, which concentration depends on atmospheric conditions. The quantification based on thermochemical predictions is globally accepted.

Aluminum oxide particles The knowledge about aluminum oxide particles is quite advanced, but rather sparse. Particle size distribution was obtained from sub-scale motor firings, post-firing ground collection, impingement on surfaces or even direct collection from post-launch wakes through aircrafts. Empirical correlations based on a wide database connecting operating pressure, propellant composition, and throat area were published by Hermsen.[29] Recently, Carlotti and Maggi published the results of a direct collection from supersonic plume with a dedicated methodology. The technique enabled detailed microscopic analysis, particle size distribution measurement, and chemical characterization.[30,31] Some data variability can be observed among different authors, depending on measurement technique and rocket motor type. Ejected particles span from sub-micrometric range to about 20 micron.[32,33] Other authors find smaller sizes, such as Strand and co-authors who found only particles in the sub-micrometric range[34] or Carlotti and Maggi who identified particles up to few micrometers.[31] The shape of the smallest condensed combustion products appear solid spherical while hollow particles can be observed for the larger fraction. X-Ray diffraction analysis unveiled that most of particle composition is made by gamma-alumina, being also alpha-alumina present.[35] *In-situ* measurements of Athena II plume at 18 km altitude showed that most of the particles (up to about 99% by mass) were large enough to have a short lifetime in the atmosphere.[11]

Soot At author's knowledge there are not specific studies about soot emission from solid rocket motors powered with aluminized propellants. These energetic materials operate an oxygen-lean combustion. These condition may favor the generation of carbon residues. Traces were found in collection data by Carlotti and Maggi but it is not clear whether these residues resulted from a contamination originated by the experimental apparatus.[31] Conversely, spectroscopic analyses by Kolz and co-authors performed on the flame of AP/HTPB propellant strands did not reveal the presence of soot.[36] According to the present knowledge, it seems that carbon black release may be of lesser importance for aluminized solid propellant rocket motors.

Nitrogen carriers Nitric acid was identified by plume probing through high-altitude aircraft. Non-negligible concentrations were found for reactive nitrogen ($NO_x$) and for nitric acid ($HNO_3$) in a 30-minute old plume of an Athena II SRB.[11]

### 5.2. Hydrocarbon/oxygen liquid propulsion units

In this category we can find both the typical RP-1/LOX and the novel CH4/LOX. The former one is a well known propellant couple. The latter one represents a cryogenic candidate for future propulsion units, competing with hydrogen/oxygen in terms of mean density, storage temperature, at the cost of a limited specific impulse loss.

Soot Rockets powered by RP1 fuel are known to generate soot. Its formation depends on several parameters such as OF ratio, injection pattern, combustion pressure, .... Soot load in exhaust plume is uncertain as some oxidation occurs after the mixing with the atmosphere. Emission indexes used for climatological predictions are often based on estimates, having scarce availability of dedicated experimental data. Experimental data are based on spectroscopy. Plastinin and co-authors presented a conference paper regarding the plume of an Atlas III launcher, at 18 km of altitude, using both plume radiation and sunlight scattering. They estimated a plume content of about 0,17% by mass and a particle size of 92 nm.[37] The observations are in line with typical intervals but uncertainties are still high.[38]

CH4/LOX compositions benefit from methane tendency to burn without soot. Some experimental data from spray injection were performed at DLR. Methane/oxygen combustion generated soot only under extreme fuel-rich conditions whereas no spectroscopic signature of carbon particulate is observed under typical OF ratios.[39] This propellant couple is currently under the spotlight as several propulsion units are under development so it is likely that several data become soon available.

### 5.3. Hybrid rockets

From the experimental viewpoint, hybrid rocket plume characterization lacks of fundamental research activity. In a recent paper a measurement of soot concentration is published by Aphale and co-authors.[40] The paper considers a slab burner operating at ambient pressure. Experimental concentrations range between about 3 to 9 ppm of soot along the 1-cm axis of the fuel grain. However, in real rocket configurations further oxidation in post-chamber may occur, reducing the effective exhaust and making it sensitive to configuration and operating conditions. Systematic studies are missing.

### 5.4. Other aspects

One important aspect is correlated to the use of storable oxidizers based on nitrogen. Nitric acid, nitrous oxide, or hydrazine are examples of molecules which can contribute to active nitrogen generation. At author's knowledge, nitrous oxide effect is not documented. On the hypergolic couple UDMH/N2O4 Ross and co-authors considered in their model NO and water vapor as main contributors to ozone depletion. A Proton launch was analyzed and results predicted a more limited global impact, when compared to chlorine-based depletion processes. The fact was attributed to a different reaction kinetics of the two radical processes and to a different (assumed) emission index.[41] The authors also added an important aspect about soot and correlated uncertainties. UDMH engines tend to develop lower amount of soot with respect to RP1-based systems but vigorous afterburning consumes most of the carbon particles in the latter. This process is not present in UDMH engines.

## 6. Final remarks

This paper revised sets of literature information about the environmental impact of space launchers. From main mechanisms of plume interaction with the atmosphere, most important rocket emissions have been analyzed. Finally, considerations and experimental characterization data about plume features for most important rocket propulsion have been reported. This review is far from being complete, given the variety of the subject.

From a historical perspective, the knowledge of the interaction mechanisms between plume and atmosphere has been progressively refined, enabling the identification of the areas of main concern. More recently, accurate models capable of predicting the climatological effect of fleets of launchers became available. However, these predictions ground on uncertain emission indexes. It appears that systematic analysis of plume emission is currently missing. Most of the available data refer to peculiar cases and architectures, missing a general description.

The question to answer is still the same, since the late nineties: is the future space launch activity sustainable, when compared to the global human activity? Are we overlooking or are we stressing too much the problem? Finally, is the plume the most important aspect to quantify the environmental impact of a launcher? For sure, the trail of smoke behind a rocket is the most visible trace but production, disposal, and propellant supply are also part of the game.

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