

1 **Metallic and molecular ions in the magnetosphere**

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20

21 **Abstract**

22 Observations and present knowledge of heavy ions with mass ≥ 27 in the
23 magnetosphere are reviewed. There are four ultimate sources of these heavy ions: the
24 solar wind (mainly high charge-state atomic ions), the ionosphere (mainly molecular ions),
25 the atmospheric metal layers that are originating ultimately from ablation of meteoroids
26 and possibly space debris (low charge-state metallic ions and metal-rich molecular ions),
27 and lunar surface and exosphere (low charge-state metallic and molecular ions).

28 The upstream heavy ions (solar wind origin and lunar origin) give independent
29 information on the ion entry route to the magnetosphere from proton (H^+) and alpha
30 particles (He^{++}): with similar mass-per-charge (m/q) values, or gyroradius, for the solar
31 wind origin, and much larger gyroradius for the lunar origin. The lunar origin ions also give
32 independent insights from laboratory observations on the sputtering processes. The
33 atmospheric origin molecular and metallic ions are essential in understanding energization,
34 ionization altitudes, and upward transport in the ionosphere during various ionospheric and

35 magnetospheric conditions. These ions are also important when considering the evolution
36 of the Earth's atmosphere on the geological timescale.

37 Only a few terrestrial missions have been equipped with instrumentation dedicated to
38 separate these molecular and metallic ions, within only a limited energy range (cold ions of
39 < 50 eV and energetic ions of ~ 100 keV or more) and a limited mass range (mainly ≤ 40
40 amu). This is far too limited to make any quantitative discussion on the very heavy ions in
41 the magnetosphere. For example, we cannot dismiss any of four possible sources with the
42 existing data.

43 Under this circumstance, it is worth to re-examine, using available tools, the existing
44 data from the past and on-going missions, including those not designed for the required
45 mass separation, to search for these ions. The purpose of this review is to summarize the
46 availability of these datasets and tools. This review also shows some examples of
47 combinations of different datasets that provide important indications of the sources of
48 these heavy ions and their amounts that have been overlooked to date.

49 Finally, we note the possible future contamination of specific masses (mainly Aluminium
50 (Al), but also lithium (Li), iron (Fe), nickel (Ni), copper (Cu), titanium (Ti) and germanium
51 (Ge)) by the ablation of re-entering human-made objects in space (debris and alive
52 satellites) in the coming decades. This possibility argues the need for dedicated
53 observations of magnetospheric and ionospheric metallic ions before these metallic ions of
54 space debris origin start to dominate over the natural contribution. The required
55 observations can be performed with the available designs of space instrumentation and
56 available ground-based instruments.

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58

59 **Acronyms**

60 CIR: Co-rotating interaction region

61 CME: Coronal mass ejection

62 ESA: electrostatic analyzer

63 ESA: European Space Agency

64 FOV: field of view

65 IMF: interplanetary magnetic field

66 IS radar: incoherent scatter radar

67 LEO: low-Earth orbit

68 lidar: light detection and ranging (now is accepted as normal noun, though)

69 MCP: microchannelplate

70 PHA: pulse height analysis

71 QL: quick look plot

72 RPA: retarding potential analyser

73 SSD: solid-state detector

74 TOF: time of flight

75

76 Space missions and instruments

77 ACE: Advanced Composition Explorer

78 ULEIS: Ultra-Low-Energy Isotope Spectrometer

79 AE-C and AE-D: Atmosphere Explorer C and D

80 AMPTE: Active Magnetospheric Particle Tracer Explorers

81 CCE/CHEM: Charge-Energy-Mass Spectrometer on board Charge Composition Explorer

82 IRM/SULEICA: SUprathermaL Energy Ionic Charge Analyzer on board Ion Release Module

83 ARGOS/ISAAC: Ionospheric Spectroscopy and Atmospheric Chemistry instrument on board the

84 United States Air Force's Advanced Research and Global Observing Satellite

85 ARTEMIS: Acceleration, Reconnection, Turbulence and Electrodynamics of the Moon's Interaction

86 with the Sun

87 ASAN: Advanced Small Analyzer for Neutrals on board Chang'e-4

88 au: Astronomical unit

89 Cassini/MIMI-CHEMS: Charge-Energy-Mass Spectrometer of Magnetosphere Imaging Instrument

90 on board Cassini

91 Chandrayaan-1/SARA: Sub-keV Atom Reflecting Analyser

92 Cluster/CIS/CODIF: COmposition DIstribution Function of Cluster Ion Spectrometry

93 CRRES: Combined Release and Radiation Effects Satellite

94 DE: Dynamics Explorer

95 RIMS: Retarding Ion Mass Spectrometer

96 EICS: Energetic Ion Composition Spectrometer
97 Geotail:
98 STICS: Supra-Thermal Ion Composition Spectrometer (STICS)
99 LEP: Low Energy Particle experiment
100 ISIS: International Satellites for Ionospheric Studies
101 JUICE/PEP: Particle Environment Package on board JUpiter ICy moons Explorer
102 JDC: Jovian Dynamics and Composition
103 NIM: Neutral gas and Ion Mass spectrometer
104 LADEE: The Lunar Atmosphere and Dust Environment Explorer
105 Kaguya/IMA: Ion Mass Analyzer of MAgnetic field and Plasma experiment - Plasma energy Angle
106 and Composition Experiment (MAP-PACE) on board Kaguya
107 MAVEN/STATIC: SupraThermal And Thermal Ion Composition on board Mars Atmosphere and
108 Volatile Evolution mission
109 MMS/HPCA: Hot Plasma Composition Analyzer on board Magnetospheric MultiScale mission
110 OGO: Orbiting Geophysical Observatory satellite
111 STEREO/PLASTIC: Plasma and Suprathermal Ion Composition on board Solar Terrestrial
112 Relations Observatory
113 SOHO/CELIAS: Charge, Element, and Isotope Analysis System on board SOLar and Heliospheric
114 Observatory
115 VAP: Van Allen Probes
116 Wind STICS: Geotail:
117 STICS: Supra-Thermal Ion Composition Spectrometer (STICS)
118
119 Models
120 CABMOD: Chemical Ablation Model
121 LEGEND: LEO-to-GEO Environment Debris model
122 MASTER: Meteoroid and Space Debris Terrestrial Environment.
123 ORSAT: Object Reentry Survival Analysis Tool
124 SAMI: Southern Appalachian Mountains Initiative
125 SCARAB: SpaceCraft Atmospheric Re-entry and Aerothermal Break-up
126 WACCM: Whole Atmosphere Community Climate Model
127 WACCM-X: WACCM with thermosphere and ionosphere eXtension
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130 **1. Introduction**

131 Heavy ions have been found in the magnetosphere since the early 1970's (Shelley et
132 al., 1972; Hoffman et al., 1974). Subsequently, magnetospheric ion dynamics has been
133 studied with four major components (H^+ , He^{++} , He^+ , and O^+). Both helium and oxygen have
134 been used as markers for plasma coming from the solar wind (He^{++}), plasmasphere (He^+),
135 and ionosphere (O^+), respectively, while we cannot distinguish the origin of H^+ . The minor
136 components thus give important information regarding the source and transport.

137 This applies to the even less-abundant minor ions such as molecular ions and metallic
138 ions. The molecular N_2^+ , NO^+ , or O_2^+ ions, compared to the atomic O^+ ions (without
139 separating from N^+), carry certain important information on the internal processes in the
140 ionosphere as well as the ion outflow processes above the ionosphere. The metallic ions
141 give information on the metal deposit to the mesosphere, mesosphere-thermosphere
142 coupling, as well as on the lunar-origin ions and its entry route to the magnetosphere.
143 Such information is not obtained from the four major ion species (H^+ , He^{++} , He^+ , and O^+).

144 For example, the lunar origin ions of four major species are completely masked by
145 those of solar wind origin. Similarly, H^+ , He^+ , and O^+ originating from the lower part of the
146 ionosphere and mesosphere are masked by those originated at upper ionosphere. Thus,
147 the heavy molecular and metallic ions provide unique information that is not obtained from
148 the four, and contributes to understanding many aspects of the geospace environment: the
149 ion escape process from the ionosphere, related ionospheric and even mesospheric
150 process, deposition of near-Earth small bodies at different altitudes, solar wind-
151 magnetosphere interaction, and even the dynamics of the Moon-origin ions.

152 The molecular and metallic ions were actually found a long time ago by the ISIS-2 and
153 DE-1 satellites (Hoffman et al., 1974, Chapell et al., 1982, Craven et al., 1985). Despite
154 this, the roles of such heavy ions ($mass \geq 27$) and their composition in the terrestrial
155 magnetosphere have received little attention, and the observations of heavy ions and
156 molecular ions in the magnetosphere are sparse. The limited numbers of existing
157 magnetospheric observations are often the bi-products from missions with other prime
158 objectives.

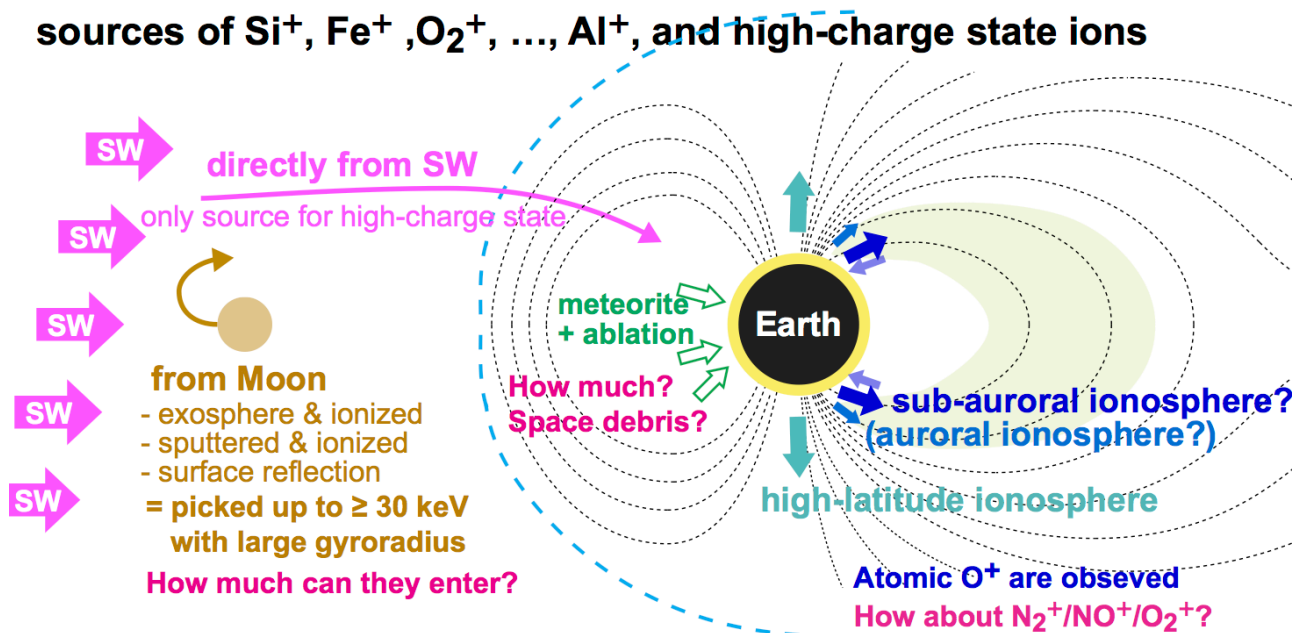
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160 **1.1. Four possible sources**

161 For heavy molecular ions, the majority is of ionospheric origin, and supplied mainly from
162 the polar ionosphere (e.g., see review by Lin and Ilie, 2022), but there may be

163 contributions from other sources and via other routes. For the metallic ions with energies
 164 greater than 100 keV/q (energy range of Supra-Thermal Ion Composition Spectrometer
 165 (STICS) instruments on board Geotail and Wind spacecraft), the majority are high charge-
 166 state solar wind ions such as Fe^{12+} , while some ions are low charge-state such as Fe^+ and
 167 K^+ (e.g., Christon et al., 2017). Considering the ion distribution of the topside ionosphere
 168 and its further energization to reach the magnetosphere against the escape energy, the
 169 supply of metallic ions from the ionosphere to the magnetosphere is expected to be very
 170 small. This makes it difficult to understand how these metallic ions are provided to the
 171 magnetosphere and where they come from.
 172

sources of Si^+ , Fe^+ , O_2^+ , ..., Al^+ , and high-charge state ions



174 Figure 1.1. <metallic_source>: Illustration of possible sources of heavy molecular ions and
 175 metallic/silicate ions to the magnetosphere. Solid arrows show the pathway of these ions:
 176 (1) solar wind for high charge-state metallic/silicate ions (we call them simply "metallic
 177 ions" hereafter), (2a) dayside polar ionosphere (cusp and its vicinity) low charge-state
 178 heavy ions, (2b) night-to-morning auroral and sub-auroral ionosphere molecular ions, (3)
 179 mesospheric layer of heavy elements, which are ultimately provided by meteoric ablation,
 180 for metallic/silicate ions, and (4) The Moon surface and exosphere for low charge-state
 181 heavy ions. Note that the outflow mechanism from mesospheric layer means the atoms or
 182 ions must receive the same electromagnetic energization mechanisms as ionospheric
 183 ions.
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 186 As illustrated in Figure 1.1, there are four major sources of heavy ions in the Earth's
 187 magnetosphere with mass > 27 amu (here, we limit our discussions to mass < 100 amu,
 188 i.e., molecules, metallic atoms and their ionized forms). These sources are: (1) The solar

189 wind; (2) The ionosphere (and thermosphere); (3) The mesospheric layer of heavy
190 elements (hereafter, "metal layers") for which the metallic ions have been detected by the
191 sounding rocket since 1970; and (4) The surface and exosphere of the Moon. Here, the
192 upper atmospheric sources are further divided into: (2a) the dayside polar ionosphere
193 (cusp and its vicinity) low charge-state heavy ions, and (2b) night-to-morning auroral and
194 sub-auroral ionosphere molecular ions. Unlike the molecular ions (N_2^+ , NO^+ and O_2^+) that
195 can be produced in the upper part of the ionosphere, metallic ions come from (3)
196 mesospheric metal layers because of the low concentration of metallic atoms in the
197 atmosphere otherwise.

198 The mesospheric metal layers are produced by mainly the ablation of cosmic dust
199 (meteoroids), but a possible contribution from the ablation of space debris may increase in
200 near the future because of the rapid increase of space debris and satellites. The ablation
201 of these human-made objects during their atmospheric re-entry from space (in this paper
202 termed as "ablation of space debris/waste") results in the deposition of human-made
203 metallic atoms to the mesosphere. Some common metals in spacecraft such as aluminum
204 (Al) and lithium (Li) are very rare in the natural meteoroids, and may substantially
205 contribute to the metallic layers, which in the end contribute to the magnetospheric metallic
206 ions.

207 While the high charge-state ions and majority of the molecular ions come from (1) the
208 solar wind and (2) the high-latitude ionosphere, respectively, the sources of the low
209 charge-state metallic ions (including Si^+ , and we simply refer them "metallic ions"
210 hereafter) have not been identified. Two theoretically possible candidates are (3) the
211 mesospheric metal layer and (4) the Moon, but both the sources have a too-low flux
212 problem, and hence the possibility of coming from upstream solar wind is not dismissed.
213 So far, not a single satellite observation has confirmed (or even indicated) the source and
214 condition.

215 Here, we cannot dismiss the Moon as a source of metallic and molecular ions
216 (Hilchenbach et al., 1993; Mall et al., 1998), as the result of sputtering at the surface (Saito
217 et al., 2010) or photoionization in the thin exosphere (Halekas et al., 2015). When the
218 Moon is upstream of the Earth (near the new Moon), these sputtered and photoionized
219 ions are picked up by the solar wind plasma and gain up to twice the solar wind velocity
220 (tens keV to hundred keV), with sufficiently large gyroradii to penetrate the
221 magnetosphere. The problem is their flux. The initial result of Apollo did not show the
222 upper lunar atmosphere containing metallic species, and only upper limits have been

223 derived from ground-based observations (Stern, 1999). Although Kaguya found a
224 signature of metallic ions at 100 km altitude around the Moon by in-situ measurement
225 (Saito et al., 2010), and the Lunar Atmosphere and Dust Environment Explorer (LADEE)
226 confirmed the thin exosphere with heavy species (Poppe et al., 2016b), the Moon-origin
227 ion flux in the Earth' magnetosphere must be nearly impossible to detect after spreading
228 over the large volume of the magnetosphere.

229 The very low ion flux also applies to the mesospheric source. Although the upward ion
230 convection below the exobase is common and the escape energy is similar (about 20 eV
231 for mass 30) between the metallic atoms (or ions) and molecules (or molecular ions), we
232 expect the metallic ion flux at the exobase to be much lower than that of the molecular
233 ions. The heavy mass (high escape energy) also makes it difficult for metallic ions to reach
234 the ion energization region above the exobase where ions are accelerated to more than
235 the escape energy. In the best case when any metallic ions are upflowing from the
236 exobase, it must be in the polar region where the magnetic field line is near vertical and
237 upflowing molecular ions with similar masses are observed (Yau et al., 1993). Note that
238 this energization works for ions but not for neutrals. Since the density ratio of ion to
239 neutral is small at the lower part of the ionosphere, drag of neutrals by the ions is not very
240 effective to gain the escape energy at exobase, and therefore, we hereafter focus on ions
241 but not neutrals.

242 These possible sources for low charge-state metallic ions, in the best case, may
243 account only for a very low flux in the magnetosphere. On the other hand, such low flux
244 that makes detection difficult, may enable these ions to work as a good marker of how
245 these ions are transported from the source. Considering the completely different energy
246 and energy/charge from the light ions or high charge-state ions (large gyroradius and high
247 energy), the distribution and amount of these metallic ions give independent information
248 on the entry of ions from the solar wind and from the ionosphere. Thus, these very minor
249 ions work as good tracers.

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252 **1.2. Sciences related to each source**

253 The source mechanism of metallic or heavy molecular ions (producing them and
254 bringing them into the magnetosphere) are different between different sources. This opens
255 up a possibility of classifying these ions in the magnetosphere in terms of dependency on
256 the source condition (which also depends on the geomagnetic and solar wind conditions).

257 Together with the fact that they are very minor in the magnetosphere, quantitative
 258 measurements of these ions in the magnetosphere through each source at various places
 259 and various condition help revealing the source mechanisms of various source, and thus
 260 contribute to many science topics, as summarized in Table 1.1.

261 First, metallic (and heavy molecular) ions coming from the ionosphere carry essential
 262 information of the particle transport (by electromagnetic and electrostatic fields) in a
 263 collision-free frame from the exobase, particle transport in the ionosphere (and even in the
 264 mesosphere), ionization and chemistry in the ionosphere, and dynamics of the metal
 265 layers in the mesosphere and ionosphere. Here, ablation of the cosmic dust and near-
 266 Earth small bodies is considered as the source of the metal layers, and hence metallic ions
 267 of mesospheric and thermospheric origin are largely attributed to meteoroids.

268 In addition, the amount of escaping molecular ions gives extra information on the
 269 evolution of the Earth's atmosphere, although the main evolution is caused by the surface
 270 interaction including biological activity. Here, the evolution is not limited to the change in
 271 the total amount of oxygen, but also includes the oxidization rate (low CO₂/O₂ ratio and low
 272 N₂/O₂ ratio compared to Mars and Venus) and its fluctuation (just 5% change in 0.1 million
 273 scale often happen). The observed O⁺ escape suggests a substantial contribution the
 274 latter (Yamauchi, 2019; Dandouras et al., 2018; 2016).

275

276 Table 1.1: Possible scientific contribution of heavy ion measurement

Ion source	Relevant science topics
(1) Solar wind (metallic ions)	Solar wind - magnetosphere interaction for different masses (with similar E/q) Ion entry into the magnetosphere
(2) Ionosphere (molecular ions)	Magnetosphere-Ionosphere coupling
	Energization processes at low altitude
	Ionospheric chemistry and dynamics for both dayside and nightside
	Atmospheric evolution (high latitude source only)
(3) Mesosphere / lower thermosphere (metallic ions)	Thermospheric dynamics
	Expansion of mesosphere and mesosphere-thermosphere- ionosphere coupling
	Ionospheric dynamics on both dayside and nightside
	Flux and deposition altitude of meteoroids
	Ablation and deposition of space debris with lower velocity than meteoroids
	On-orbit space debris fragmentation
(4) Moon surface and exosphere (low charge-state ions)	Solar wind - surface interaction different mass (with similar E/q)
	Micrometeorite-surface interaction
	Solar wind-magnetosphere interaction for pick-up heavy ions with different masses (with different E/q)

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279 The low charge-state metallic ions in the magnetosphere can also be a possible
 280 indicator of space debris because, in the near future, we expect a substantial increase of
 281 human-made heavy atoms/ions (e.g., Lithium (Li) with mass 7, Aluminum (Al) with mass
 282 27, Silicon (Si) with mass 28, Titanium (Ti) with mass 48, Iron (Fe) with mass 56, Nickel
 283 (Ni) with mass 59, Copper (Cu) with mass 64, and Germanium (Ge) with mass 73)
 284 deposited to the mesosphere during the ablation of space debris/waste as mentioned
 285 above. Among them, those relatively rare in the natural meteoroids may become
 286 detectable. These amounts, although they are small, might exceed the natural heavy ions
 287 in the near future if no new countermeasures against space debris are taken before the
 288 Kessler syndrome (Kessler and Cour-Palais, 1978) starts over a wide altitude range (which
 289 might have started at a limited altitude around 500-800 km in the worst-case estimate).

290

291 Table 1.2: Overview of heavy ion observations in the terrestrial magnetosphere.

Species	Mass – Energy range	dedicated or not	Examples (Earth/Moon missions)
C ⁺ , N ⁺ , or O ⁺ (m/q=14–16)	altogether (not distinguished)	dedicated*1	Cluster, VAP, MMS, etc
	each of C, N, O (<50 eV)	non-dedicated*2	DE-1, Akebono
	each (0.05–100 keV)	(missing)	
	each (>100 keV)	non-dedicated	AMPTE, Geotail
N ₂ ⁺ , NO ⁺ , or O ₂ ⁺ (m/q=28–32)	altogether (<50 eV)	dedicated	e-POP, ISIS-2
	altogether (10–100 keV)	dedicated	Arase
	altogether (>100 keV)	dedicated	CRRES, Polar
	each (<50 eV)	non-dedicated	DE-1, Akebono
	each (0.05–10 keV)	(missing)	
	altogether (0.05-30 keV)	non-dedicated	Cluster, Arase, MMS, etc
	each (30–100 keV)	non-dedicated	AMPTE
	each (>100 keV)	non-dedicated	AMPTE, Geotail
High charge-state heavy	each (>100 keV)	dedicated	Geotail, Wind, AMPTE, etc
Low charge-state metallic	each (>100 keV)	dedicated	Geotail, Wind
	each (cold)	dedicated@moon	LADEE
	each (0.01–30 keV)	dedicated@moon	Kaguya, LADEE

292 *1. Routine, *2 Non-routine,

293

294 Finally, the lunar origin low charge-state metallic ions give information of how the ions
 295 with very large gyroradii are picked up and enter the magnetosphere (through the bow
 296 shock, magnetosheath, and magnetopause). The entry processes and routes must be
 297 quite different from those of the high charge-state metallic ions originating from the solar
 298 wind and the solar energetic particles. To investigate them, we need to monitor the lunar

299 origin ions near the Moon, which also allows us to further understand the sputter process
300 on the lunar surface and striving process of the lunar exosphere for different solar wind
301 conditions including its composition.

302

303

304 **1.3. Lack of observations**

305 Unfortunately, these heavy ions and atoms are largely unexplored in near-Earth space,
306 and no terrestrial mission has had a dedicated instrument for the mass range covering up
307 to the metallic ions at energy less than 100 keV, as summarized in Table 1.2. Past and
308 recent works in detecting these molecular and metallic ions were carried out with non-
309 dedicated instruments, such as plasma instruments designed to separate only four major
310 species H^+ , He^+ , He^{++} , and O^+ . Thus, the current understanding is at a preliminary level,
311 and in the best case, only qualitative (even for the major four possible sources in Figure
312 1.1).

313 Due to such difficulties in extracting the heavy molecular and metallic ions from the
314 data, past works on these very heavy ions in the magnetosphere are very sparse,
315 spanning over many years by different missions. Furthermore, it is difficult to have event
316 studies between different spacecraft, because the observation of metallic ions requires a
317 long integration time (to accumulate adequate counting statistics for these low-abundant
318 species), and active observations of wide mass ranges between different missions do not
319 normally overlap within the same time period. With these reasons, obvious question
320 remains such as whether any source is dominant over other sources, and if so, under what
321 conditions. At the present, even the relative importance of ions of the Moon origin and
322 Earth origin of different species is not known.

323 Any quantitative (statistically significant) measurements in the future will give important
324 information on the transport of these very heavy ions in the magnetosphere: e.g., on the
325 entry route, acceleration, and dependence on the external condition. On the other hand,
326 during the best conditions such as large geomagnetic storms, some of the existing
327 instruments on-board the past and current spacecraft listed in Table 1.2 actually separated
328 N^+ , O^+ , N_2^+ , NO^+ in observations, and even metallic ions (Fe^{n+}) after sufficient integration
329 of the data over times. In other words, existing data from past and on-going missions are
330 capable of detecting them, although the result is qualitative. Thus, it is very useful to make
331 a list of all possible instruments that can potentially detect heavy molecular and atomic
332 ions. Of course the limitation is severe. Even tracing the source of the observed metallic

333 ions is often not possible from the available data except for some obvious cases such as
334 near the interaction region with the solar wind.

335 In the list, we should include the Moon and solar wind missions that actually observed
336 the Earth's magnetosphere, if the mission included a mass spectrometer better suited for
337 these investigations than those onboard magnetospheric missions. One example is the
338 Kaguya Moon mission that was inside the magnetotail or upstream of the magnetosphere
339 for substantial time periods (Saito et al., 2010). Another example is the WIND mission that
340 traversed the magnetotail for many hours. Non-mass resolved ion spectrometers with a
341 wide energy range might also detect heavy ions when ions are picked up and flowing with
342 the solar wind velocity, such as the observation of Venus downstream at the Sun-Earth
343 Lagrange point (Grünwaldt et al., 1997). Ideally, Earth-flybys of planetary missions should
344 also be included because they normally carry mass spectrometers with much better mass
345 resolution at energies of 0.01–100 keV than those on magnetospheric missions, but it is
346 not very practical to include them because very limited data are taken by only Cassini and
347 STEREO.

348 Since the molecular and low charge-state metallic ions were not a prime observation
349 target of the all past and current magnetospheric missions listed in Table 1.2, software
350 tools and methodology to extract information about these heavy ions are normally not well
351 documented or maintained. Thus, there is a risk of losing the relevant data (including the
352 case that the data becoming unreadable with modern computer systems) even for data
353 that are used as examples of the reported metallic or heavy molecular ions. Therefore,
354 collection of the capable missions and instruments requires information of software
355 availability. This is very difficult for terminated missions if not impossible.

356 It would be also useful to combine different types of observations, e.g., solar wind
357 monitor, Moon data, satellite data in both the magnetosphere and ionosphere, and ground-
358 based (lidar) data. We present several examples that can actually update the present
359 knowledge about the heavy ion entry into the magnetosphere.

360 Finally, we discuss a possible “ideal” and “compromised” suite of observations,
361 including feasible improvements of the instruments (including software). Such a suite helps
362 planning the future observation by many capable missions (such as onboard the Space
363 Safety/Earth Observation/Lunar Exploration missions) as well as a dedicated mission.
364 Here, we aim to answer fundamental questions that arise for different conditions, such as:

365

- 366 - What is the relative importance of the terrestrial (ionosphere or mesosphere) source
 367 and the lunar source for the low charge-state heavy ions in the magnetosphere?
 368 - How much of the observed heavy ions emanate from the aurora/sub-auroral
 369 ionosphere compared to the cusp and its vicinity?
 370 - Is it possible to quantitatively evaluate the present and future contamination by
 371 space debris?

372

373 The review has the following sections.

374

375 2. The scientific importance of molecular and metallic ions

376

377 2.1 Importance and present knowledge of the solar wind source (high charge-state metallic
 378 ions)

379 2.2 Importance and present knowledge of the ionospheric source (molecular ions)

380 2.3 Importance and present knowledge of the mesospheric sources (metallic ions)

381 2.4 Importance of the Moon as source and present knowledge (sputtering and pickup
 382 process)

383

384 3. List of datasets and models that actually detected molecular/metallic ions

385 3.1 Satellite datasets

386 3.2 Analyses tools to extract molecular/metallic ions in the space missions

387 3.3 Mesospheric and ionospheric dataset including sounding rocket

388 3.4 Modelling of contribution from meteor and space debris through deposition to the metal
 389 layer

390

391 4. Merit of combining data from different sources and models

392 4.1 Moon contribution to energetic (> 100 keV) ions

393 4.2 Ionospheric origin of low-energy heavy molecular ions (in-situ observations)

394 4.3 Upper atmospheric source of metallic ions (Ground-based observation, Model)

395 4.4. Re-entering space debris as a heavy ion source: Outlook and unanswered questions

396

397 5. Summary and Future observation

398 5.1 Summary of unanswered science questions

399 5.2 Desired specification for observation and model

400 5.3 Desired missions and observations

401 5.4 Modelling of upward transport of metallic and molecular ions in the ionosphere and
 402 mesosphere

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404 6. Conclusion

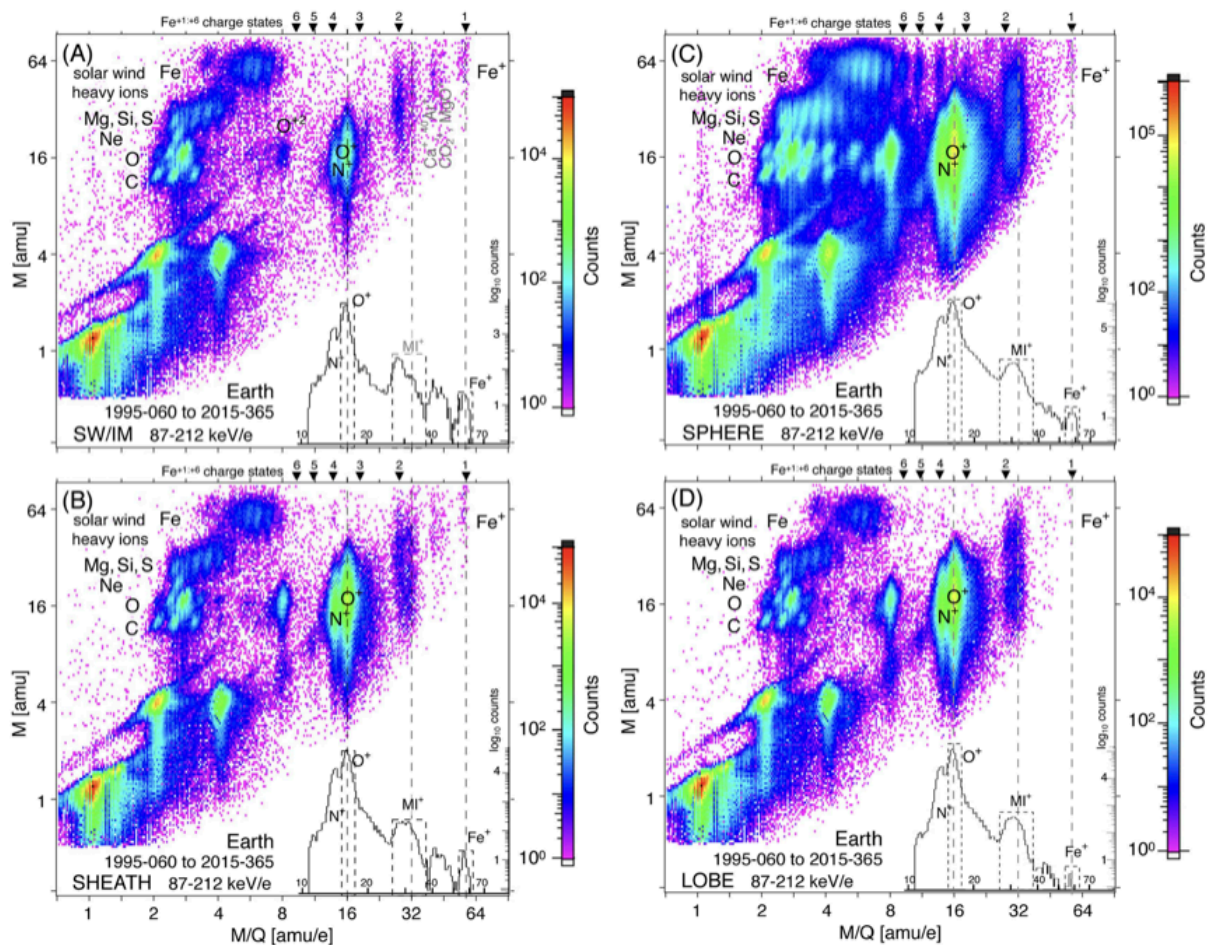
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408 **2. The scientific importance of molecular and metallic ions**

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<<Fig 2.1: Steve_100-200keV/e Ions@Earth_a>>: Average 100-200 keV/e ion composition in and near Earth's Magnetosphere. Mass – mass-per-charge ($m - m/q$) diagrams are shown for 4 different regions (see Christon et al., 2017 for definition): (a) the upstream region (SW/IM), (b) the magnetosheath (SHEATH), (c) the magnetosphere (SHERE), and (d) lobe, respectively. The counts found in the upper left half of each panel correspond to high charge-state ion of the solar wind origin. At lower right part of each panel, histogram for m/q is also shown for low charge-state heavy ions. Here, m/q is obtained from the combination of electrostatic analysis (giving energy-per-charge E/q) and time-of-flight TOF (giving velocity), and m is obtained from the combination of pulse height analysis PHAs (giving total energy E) and TOF. The measurement accuracy is the best for E/q and the worst for E , resulting in more spread in the m direction than in the m/q direction.

Figure 2.1 shows Geotail/STICS statistics of the count distributions over 20 years, ordered by mass (m) and mass-per-charge (m/q) in four different regions (Christon et al., 2017). In the magnetosphere (Figure 2.1c: SPHERE), low charge-state metallic ions (Fe^+) are seen

428 as (marked as 1 at the top of the panel) isolated from high charge-state Fe ions, in
429 addition to heavy molecular ions (MI^+) at around $m/q \sim 30$.

430

431

432 **2.1 Importance and present knowledge of the solar wind source (high charge-state** 433 **metallic ions)**

434

435 The solar wind is the obvious source of high charge-state metallic ions in the terrestrial
436 magnetosphere because the high temperature of the solar corona provides for multiple
437 high charge-state ionizations of all elements. During the travel or even at the corona, ions
438 are often energized to more than 1 MeV, forming the solar energetic particles (SEPs), and
439 these ions have sufficiently high energy to be detected by mass spectrometers of
440 energetic particles, as shown in Figure 2.1. Note that the term "solar wind" does not
441 necessarily mean that all these elements are flowing with the solar wind proton flow.
442 Some solar flare origin energetic ions take different path such as along the IMF, while
443 some are accelerated at the interplanetary shock to reach the energy detectable by the
444 Geotail/STICS instrument (Figure 2.1). Even atoms and of interstellar origin, including
445 galactic cosmic rays, are also present in the solar wind, although in much smaller
446 abundances than those of solar origin.

447

448 Figure 2.1 also shows existence of Fe^+ and Fe^{++} , isolated from high charge-state Fe ions,
449 indicating upstream source of atomic metals, i.e., other than the solar corona. The
450 obvious candidate is the Moon when it is upstream, as discussed in Sect. 2.4. Such low
451 charge-state metallic ions have quite different gyroradius from the solar wind protons.
452 Considering it is difficult to provide from the ionosphere means that ions, these metallic
453 ions work as the tracer as mentioned in Sect. 1.1, i.e., one can gain a clue in the pathway
454 of these ions to the Earth by knowing the distribution and amount of these metallic ions,
455 and by comparing with those of light ions. Furthermore, the distribution of the low charge-
456 state metallic ions provides the effectiveness of their energization mechanisms across bow
457 shock and magnetospheric boundaries, and even entry mechanisms to the
458 magnetosphere because any electrostatic acceleration (e.g., at the bow shock) gives
459 different acceleration efficiency from light ions even for the same mass per charge (m/q).

460

461 A major challenge is that, compared to good-quality observations and datasets in the solar
462 wind for both the solar wind energy (generally < 100 keV and < 2 keV/q) and the SEP
463 energy (up to 100 MeV), magnetospheric observations are sparse and are typically limited
464 to high energy for high charge-state ions.

465

466

467 **2.1.1. Observations in the solar wind**

468 The observations of heavy ions in the solar wind date back to the first space missions,
469 such as the Vela missions (e.g., Hundhausen et al., 1967) and the Apollo Moon missions
470 in the 1960s. The satellites in Vela series were equipped with energy per charge
471 instruments from which composition could be derived. Heavy ions, in the form of high
472 charge-state oxygen, were reported by Bame et al. (1968a,b) using observations from the
473 Vela 3 satellite. Subsequent versions of the Vela satellites carried more advanced
474 instruments, and even heavier ions like silicon (Si) and Iron (Fe) and possibly sulphur (S)
475 of various charge states were reported by Bame et al (1970).

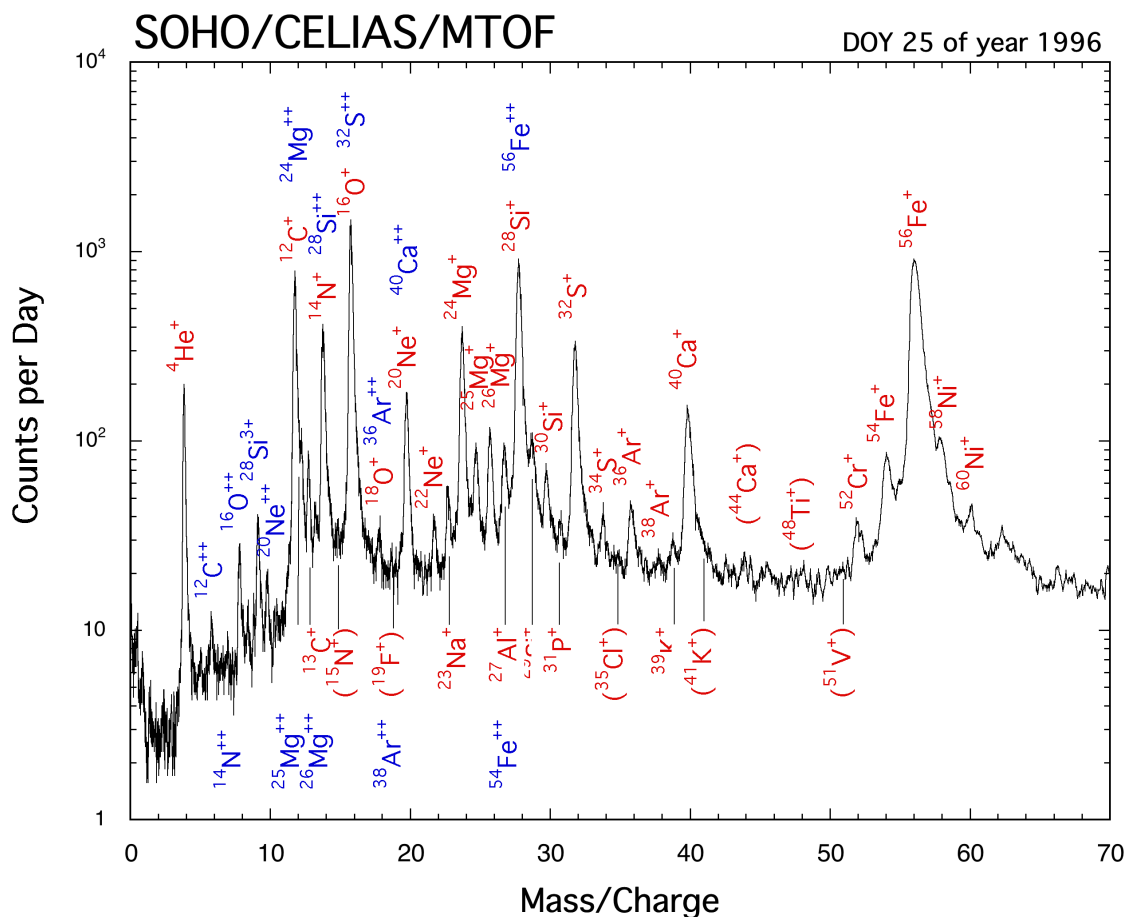
476

477 Composition of the solar wind was also inferred from depositions in the aluminium foils
478 deployed on the Moon surface by astronauts of the Apollo 11-16 missions (Geiss et al.,
479 2004) and from analysis of sample returns for updated information (e.g., Reisenfeld et al.,
480 2007; Jurewicz et al., 2007; Heber et al., 2021)

481

482 In the late 1980s and early 1990s, space missions fitted for solar wind observation such as
483 ISEE-3, Ulysses, SOHO, ACE, and WIND, measured the solar wind compositions (e.g.,
484 Gloeckler et al., 1992; Geiss et al., 1995; von Steiger et al., 2000; von Steiger and
485 Zurbuchen 2002; Reisenfeld et al., 2007). The new generation of instrumentation,
486 particularly particle spectrometers with electrostatic deflectors and time-of-flight analyzers,
487 enabled a more comprehensive mapping of the solar wind composition. For example,
488 SOHO CELIAS/MTOF instrument (Hovestadt et al., 1995) identified and classified
489 elements where no coronal spectroscopic measurements are possible (Wurz, 2005).

490



492 Fig 2.2: SOHO CELIAS/MTOF data of mass spectrum (Wurz, 2005). The charge state
 493 given in the labels refers to the charge inside the MTOF instrument (it not referring to the
 494 charge state in the solar wind), and so is the horizontal axis "Mass/Charge". Up to two
 495 main ion contributors are indicated at each peak. Labels in parenthesis indicate that the
 496 identification of that ion is highly uncertain.

497

498

499 Figure 2.2 shows a mass spectrum observed by SOHO/CELIAS with its MTOF sensor.
 500 The figure shows the variety of atomic constituents in the solar wind over a wide energy
 501 range. Table 2.1 shows an overview of metallic (atomic number $Z \geq 10$) ion species
 502 detected in the solar wind, and their abundance relative to that of oxygen (oxygen itself
 503 constitutes about 0.05–0.15% of all ions in the solar wind (e.g., Bame et al., 1975, von
 504 Steiger et al., 2010). The table even includes the solar surface data obtained by optical
 505 methods. Because many processes in the solar atmosphere, chromosphere, and corona,
 506 and the eventual formation of the solar wind depend on m/q , the composition of the
 507 photosphere and the solar wind differ by factors of 2 to 4, depending on slow and fast
 508 solar wind conditions.

509

510 Table 2.1: Relative abundance compared to Oxygen element

Element	Photosphere (*1)	Meteoritic (*1)	SW: Inter-stream (*2)	SW: Coronal-hole (*2)	SEP-derived Corona (*3)
H	1500	–	1900	820	–
Li	<0.001	<0.001	–	–	–
O	1	(*4)	1	1	1
Na	0.003	0.003	–	–	0.012
Mg	0.056	0.056	0.16	0.083	0.192
Al	0.004	0.005	–	–	0.015
Si	0.052	0.054	0.18	0.054	0.176
P	<0.001	<0.001	–	–	0.001
S	0.032	0.023	–	–	0.043
Cl	<0.001	<0.001	–	–	<0.001
K	<0.001	<0.001	–	–	0.001
Ca	0.003	0.003	–	–	0.014
Ti	<0.001	<0.001	–	–	0.001
Cr	0.001	0.001	–	–	0.003
Mn	<0.001	<0.001	–	–	0.001
Fe	0.047	0.047	0.12 - 0.19	0.057	0.224
Ni	0.003	0.003	–	–	0.008
Cu	<0.001	<0.001	–	–	<0.001
Zn	<0.001	<0.001	–	–	<0.001

511 *1: Photospheric and meteoritic abundances are from a compilation by Anders and
512 Grevesse (1989) and Grevesse and Sauval (1998)

513 *2: Solar wind abundances (SOHO/CELIAS) are taken from von Steiger (1995)

514 *3: SEP-derived coronal abundances (SOHO/CELIAS) are taken from Breneman and
515 Stone (1985).

516 *4: Assumed as the same as photosphere

517

518

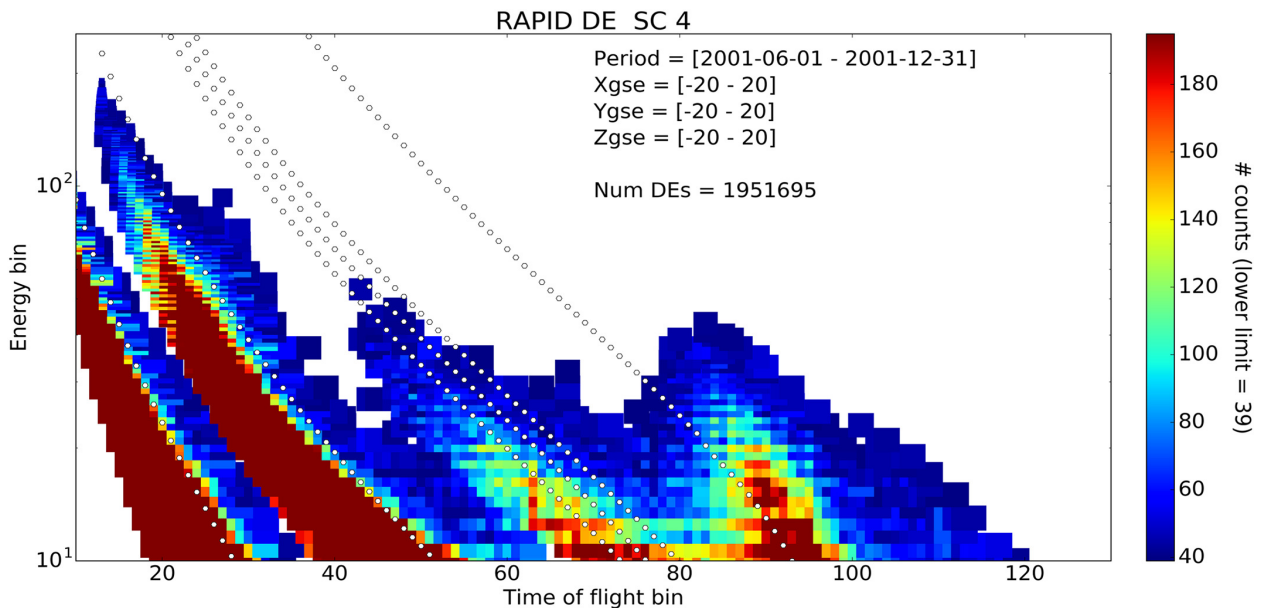
519 2.1.2. Observations in the magnetosphere

520 Observations of metallic ions in the magnetosphere are very sparse because of their high
521 mass (long TOF and large gyroradius compared to H⁺) and extremely low flux. This applies
522 to ions of both the solar wind origin (high charge-state) and the Earth/Moon origin (low
523 charge-state). Except planetary missions, only energetic particle detectors (> 100 keV that
524 give short enough time-of-flight to obtain the velocity) are capable of detecting the metallic
525 ions, such as STICS instruments on board Geotail and Wind. Ion mass spectrometers for
526 lower energies that were capable of identifying molecular ions (e.g., DE-1/EICS,
527 Akebono/SMS) did not routinely sample the full mass range. One notable exception for the
528 magnetospheric observation was the Kaguya Moon mission that could detect metallic ions,
529 although the detection of the metallic ions was limited to those from the Moon surface, but
530 not from the near-Earth region.

531

532 For the solar wind source, its contribution to metallic ions in the magnetosphere varies with
 533 the solar activity and with the solar cycle (e.g., Mitchell et al., 1983, Reames, 1995, von
 534 Steiger et al., 1997, Zurbuchen et al., 2002, Gilbert et al., 2012, Zurbuchen et al., 2016;
 535 Wurz, 2005), as well as connectivity to the Sun, e.g., slow vs. fast solar wind (e.g.,
 536 Feldman et al., 2005). Nevertheless, traces of these heavy ions are almost always present
 537 in the magnetosphere.

538



539

540 Figure 2.3: Composition measurement by Cluster/RAPID over 18 years (Haaland et al.,
 541 2020). Energy for Fe channel (no distinction of charge state) is > 400 keV, which covers
 542 higher energies than Figure 1 for Fe⁺, as well as the same energy range as solar wind
 543 Fe^{+≥7}.

544

545

546 Using AMPTE/CHEM data, Gloeckler et al. (1985) and Gloeckler and Hamilton (1987)
 547 reported that the relative abundances of the solar wind species in the magnetosphere,
 548 including high charge-state Fe ions, were very similar to the corresponding solar wind
 549 abundances, indicating that the solar wind enters the magnetosphere without significant
 550 mass discrimination. The $M - M/Q$ diagram shown in Figure 2.1.1c (Christon et al., 1994;
 551 2017) clearly show that high charge-state solar wind ions of Fe, Mg, Si, C, N, and O exist
 552 in the magnetosphere. Examining the iron charge states close to geosynchronous orbit
 553 measured by CRRES, Grande et al. (1996) showed a dramatic change of the dominant
 554 charge state from +9 to +16 during a large magnetic storm, again suggesting that the
 555 charge states measured in the outer magnetosphere reflect the changes of the charge
 556 state in the solar wind. A comparison of the iron charge states measured by ACE/SWICS

557 in the solar wind and by POLAR in the cusp/cleft region (Perry et al., 2000) showed that
558 the solar wind material direct enters during southward IMF time periods. Haaland et al.
559 (2020, 2021) identified Fe using the RAPID instrument on Cluster (which does not
560 measure charge state), as shown in Figure 2.3. They find that the abundance varies with
561 solar activity, again indicating a solar wind source. Thus, the charge state measurements
562 clearly showed that the solar wind is the dominant source of energetic iron in the
563 magnetosphere.

564

565 On the other hand, Geotail/STICS observation includes low charge-state metallic ions of
566 $m/q > 35$, such as Fe^+ , as shown in Figure 2.1. Possible sources of metals and low charge-
567 state metallic ions are the Earth's upper atmosphere, the Moon, and meteoroids, as
568 illustrated in Figure 1.1. This is a completely unexplored field because of the lack of
569 suitable instruments, even considering the very marginal capability for these
570 measurements. Therefore, when considering the magnetospheric heavy ions, we need to
571 know the charge state information even for metallic ions, although almost all signals are of
572 high charge-state of the solar wind origin.

573

574

575 **2.2 Importance and present knowledge of the ionospheric source (molecular ions)**

576

577 Molecular ions such as N_2^+ , NO^+ , and O_2^+ were found together with O^+ ions in the topside
578 ionosphere in the 1970's (Hoffman et al., 1974) and at high altitude (above $2 R_E$ altitude) in
579 1980's (e.g., Chapell et al., 1982; Craven et al., 1985). Unlike the many atomic ion species
580 in the magnetosphere (H, He, N, and O), which are partly supplied from the solar wind,
581 molecular ions cannot be of solar wind origin. In fact, Akebono SMS instrument found
582 upflowing molecular ions. One peculiar feature is that the fluxes of molecular ions (N_2^+ ,
583 NO^+ , and O_2^+) in the magnetosphere, for both the upflowing ions coming from the
584 ionosphere and trapped one in the inner magnetosphere, increase more drastically than
585 atomic ions (O^+) during strong geomagnetic storms (Yau et al., 1993; Craven et al., 1985;
586 Klecker et al., 1986; Hamilton et al., 1988). The correlation between the magnetospheric
587 activity (measured by geomagnetic indices) and the ion flux of ionospheric origin in the
588 outer magnetosphere had been recognized already in the 1970's (Geiss et al., 1978).

589

590 The drastic increase of molecular ion flux during strong magnetic storms in the inner
591 magnetosphere ($L = 2-8$) is seen over a wide energy range by the Arase satellite (Seki et
592 al., 2019; Takada et al., 2021), with the MEPI and LEPI instruments, by the velocity filter
593 effect which are capable of quantitatively separating heavy molecular ions (in the mass
594 group of 28-32) from atomic ions (Yokota et al., 2017; Asamura et al. 2018). Therefore,
595 these low-energy O^+ and O_2^+ ions in the outer magnetosphere must be, together with low
596 energy He^+ , considered to originate from the ionosphere (Young et al., 1977; Geiss et al.,
597 1978).

598

599 Since geomagnetic storms increase the energy deposition (in both forms of
600 electromagnetic energy flux and particle precipitation) from space to the ionosphere and to
601 the coupled magnetosphere-ionosphere system, we expect enhanced upward transport in
602 the ionosphere (ionospheric dynamics), enhanced production of ions in the ionosphere
603 (ionospheric chemistry), and/or enhanced pre-energization before reaching the main
604 energization region (particle dynamics above the ionosphere), as summarized in Sects
605 2.2.1, 2.2.2, 2.2.3, respectively. These effects are more drastic for molecular ions than
606 atomic ions because the threshold energy input to makes the molecular ions outflowing is
607 higher than such energy for the atomic ions. In addition, such enhancement of the
608 molecular ion outflow may influence the evolution of the life more than atomic ions, as are
609 described in Sect 2.2.4.

610

611 Although these heavy molecular ions are of ionospheric origin (lunar source is negligibly
612 small, see Sect. 2.4), the source location is not completely understood, partly because of
613 the lower fluxes compared with atomic ions and partly because of the difference in the
614 ionospheric processes between the molecular ions and atomic ions, as described in Sect.
615 2.2.3. For heavy atomic ions (O^+), there are two major outflow regions: one is the dayside
616 polar region (cusp and its downstream region) and the other is auroral region where
617 auroral acceleration is taking place (Moore et al., 1999, Peterson et al., 2008). On the
618 other hand, the Akebono satellite at more than $2 R_E$ geocentric distance found intense
619 fluxes of upflowing heavy molecular ion predominantly on the dayside (Yau et al., 1993),
620 while much less flux or zero flux of the molecular ions were detected in the nightside at
621 auroral latitudes in the observation where the O^+ upflow are observed.

622

623 Note that this O^+ - O_2^+ difference can still be attributed to the orbit and data sampling
624 coverage because we expect an equatorward shift of heavier species with respect to light
625 species in the nightside, where the magnetospheric convection is predominantly
626 equatorward during geomagnetic storm conditions, the condition when the molecular ion
627 upflow from the ionosphere is enhanced. Under any convection, heavier ions with the
628 same energy as the lighter ions are carried further downstream by the magnetospheric
629 convection compared with the lighter ions. Such mass dependency is actually observed by
630 Freja as the difference between the H^+ injection and O^+ injection in the opposite
631 hemisphere (Hultqvist, 2002; Yamauchi et al., 2005). Thus, we expect that the upflowing
632 molecular ions reach lower latitudes (e.g., the outer radiation belt) than atomic ions during
633 the upflow process, such that they travel beyond the latitude range of SMS operations at
634 Akebono altitude.

635

636 The coverage problem is not limited to Akebono observation because the ion instruments
637 on board the magnetospheric satellites are often turned off at the sub-auroral latitudes to
638 avoid the damage from the radiation belts, potentially missing the molecular ions flowing in
639 such low latitudes. Since the energization is expected to be quite different between
640 different latitudes, the outflowing path and/or the pre-energization process may be different
641 between the molecular ions and atomic ions.

642

643 It is yet unknown whether the ionospheric molecular ions outflow from the topside
644 ionosphere (or exobase) in the nightside auroral/sub-auroral region, and if so, how much
645 and in what condition do they populate the magnetosphere. These questions apply even to
646 the dayside high-latitude route. By comparing these results, we can gain more information
647 on the ionospheric processes as well as the energization processes above the ionosphere.

648

649 Molecular ions of ionospheric origin can possibly be observed deep in the magnetotail ($>$
650 $50 R_E$). Although no mass spectrometer for the magnetotail observation is capable of
651 separating O_2^+ ions from O^+ ions at energy less than 100 keV (Kaguya Moon mission with
652 a capable instrument did not look at the magnetotail flow), an ion energy spectrometer
653 with large geometric factor may in principle detect it at higher energy than atomic ions (O^+)
654 because we expect the same velocity for all species. However, in reality the energy
655 spread of O^+ is wide and O^+ energy can even be 30 times as the H^+ energy. For example,
656 the Acceleration, Reconnection, Turbulence, and Electrodynamics of the Moon's

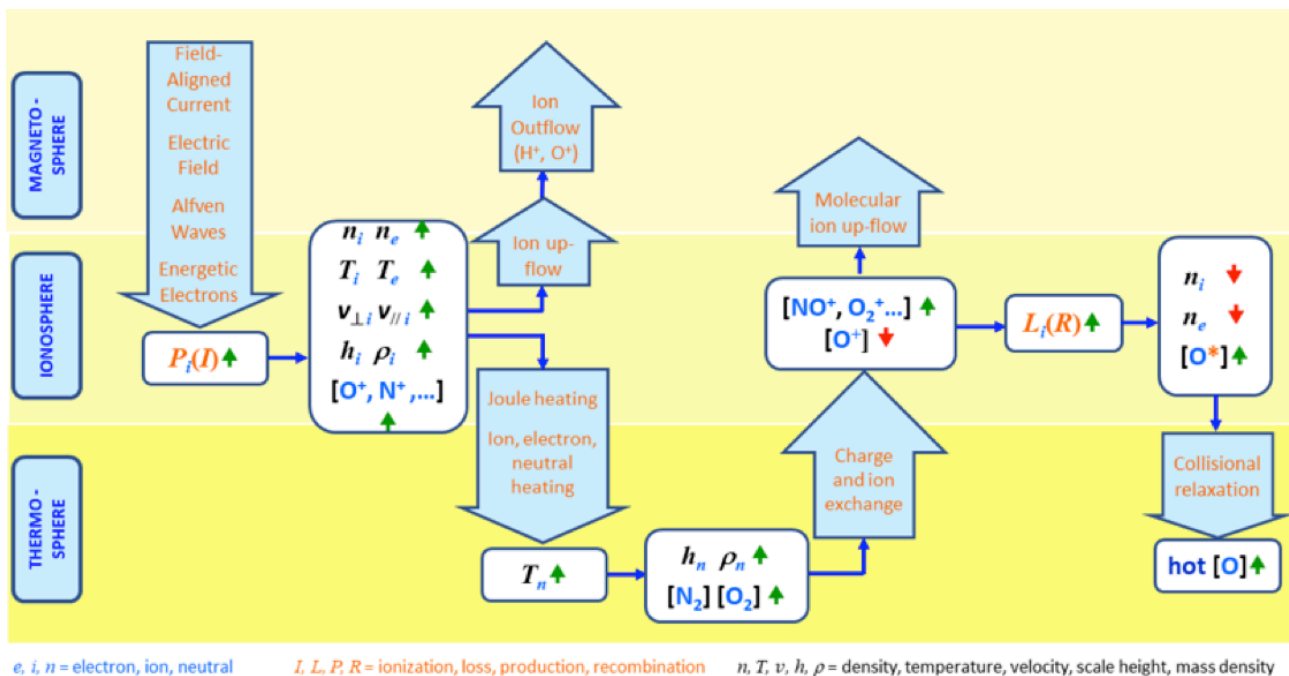
657 Interaction with the Sun (ARTEMIS) spacecraft detected an anti-sunward high ion flux at
 658 about the concurrent proton velocity, i.e., at nearly 25 times the energy as the main anti-
 659 sunward flux at the lunar distance in the magnetotail during geomagnetically disturbed
 660 times (Poppe et al., 2016a), which was initially interpreted as potentially containing
 661 molecular species (N_2^+ , NO^+ , and O_2^+ , approximately masses m/q 28–32), but majority of
 662 this high-energy flux must be atomic ions (O^+), if not all, for this event.

663

664

665 2.2.1 The importance of the ionospheric and thermospheric dynamics and 666 energization (upflow) for the molecular ions in the magnetosphere

667



668

669 Figure 2.4 \ref{fig22_Yau_outflow_schematic.png}: A schematic illustration of the
 670 underlying physical processes of ion upflow and their importance in ionosphere-
 671 thermosphere dynamics, including (a) the energy deposition in the auroral ionosphere due
 672 to field-aligned currents, convection electric fields, Alfvén waves, and auroral electron
 673 precipitation; (b) increase in ion production P_i by electron impact ionization I , increases in
 674 ion and electron densities and temperatures (n_i , n_e , T_i , T_e), ion convection and parallel
 675 velocities ($v_{i\perp}$, $v_{i\parallel}$), plasma scale height and mass density (h_i , ρ_i), and O^+ , N_2^+ , and N^+ ion
 676 composition; (c) ion upflows, Joule heating, and neutral heating; (d) resulting increase in
 677 the neutral scale height and mass density, N_2 and O densities, and N_2/O density ratio; (e)
 678 increased production of molecular N_2^+ , O_2^+ , and NO^+ ions in the F-region and above, due
 679 to increase in auroral electron impact ionization of N_2 and O_2 and in their charge-exchange
 680 and ion-exchange with the dominant O^+ ion.

681

682

683 Figure 2.4 illustrates schematically the underlying physical processes of ion upflow and
684 their importance in the ionosphere-thermosphere dynamics in the context of
685 magnetosphere-ionosphere-thermosphere (MIT) coupling, including the energy deposition
686 in the auroral ionosphere associated with field-aligned currents, convection electric fields,
687 Alfvén waves, and auroral electron precipitation (both energetic and soft electrons). Such
688 energy deposition causes an increase in ion production P_i due to electron impact ionization
689 I , and subsequently increases in ion and electron densities and temperatures (n_i , n_e , T_i ,
690 T_e), ion convection and parallel velocities ($v_{i\perp}$, $v_{i\parallel}$), plasma scale height and density (h_i , ρ_i),
691 and changes in the O^+ , N_2^+ and N^+ ion composition.

692

693 The resulting plasma pressure gradients and ion convection velocities give rise to ion
694 upflows and Joule heating, respectively, as well as neutral heating due to ion-neutral
695 collisions. The latter in turn gives rise to an increase in the neutral scale height and mass
696 density, and a corresponding increase in both N_2 and O densities at a given altitude, with
697 the N_2 density increasing at a faster rate relative to O with increasing altitude, so that the
698 increase in the N_2/O density ratio is also largest at the highest altitude.

699

700 This in turn leads to an increased production of molecular N_2^+ , O_2^+ , and NO^+ ions in the
701 ionospheric F-region and above, due to the combination of (a) auroral electron impact
702 ionization of N_2 and O_2 , both at increased densities, (b) charge-exchange of both N_2 and
703 O_2 with the dominant O^+ ion, giving rise to N_2^+ and O_2^+ , respectively, and (c) ion-exchange
704 reaction between N_2 and O^+ , giving rise to NO^+ . While contributing the significant
705 enhancement of ion upflow, these molecular ions contribute to the reduction of the overall
706 electron density due to their rapid dissociative recombination rate in the F-region, as
707 explained in details below (see also Sydorenko et al., 2016), and also contribute the
708 production of hot oxygen and nitrogen atoms in the thermosphere (see e.g., Richards et
709 al., 1994).

710

711

712 **2.2.2 The importance on the ionospheric chemistry (source and re-combination** 713 **during upflow)**

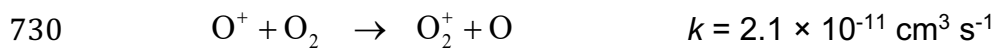
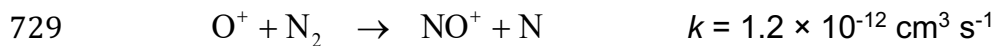
714 In terms of their influence on the ion composition in the Earth's ionosphere, the two most
715 important types of chemical processes are ion-neutral charge exchange and

716 recombination reactions. The lower thermosphere is dominated by molecular oxygen (O₂)
 717 and nitrogen (N₂), and the upper thermosphere is dominated by atomic oxygen (O) below
 718 the H-O crossover height and by atomic hydrogen (H) above, respectively.

719 Correspondingly, the E-region ionosphere is dominated by molecular NO⁺ and O₂⁺ ions
 720 (Del Pozo et al., 1997; Grebowsky and Bilitza, 2000), while the F-region and the upper
 721 ionosphere up to the crossover height is dominated by atomic oxygen ions. Above the
 722 crossover height, the topside ionosphere and beyond is dominated by atomic hydrogen
 723 ions (proton, H⁺), although the hydrogen exosphere still extends to more than 10 R_E (e.g.,
 724 Kameda et al., 2017) in the proto-dominated region.

725

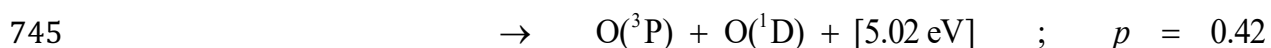
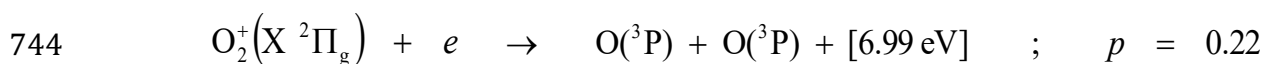
726 The charge exchange reactions of highest importance to the F-region and topside
 727 ionosphere are therefore those involving O⁺ and H⁺ ions, respectively, such as:

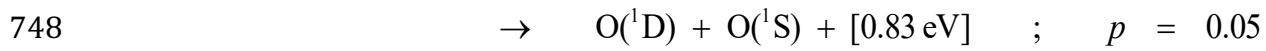
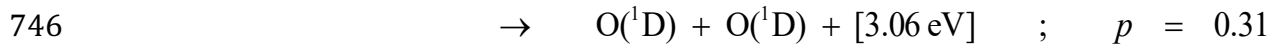


732 where k denotes the best accepted value of the room-temperature rate coefficient in the
 733 literature in each case.

734

735 Dissociative recombination processes often dominate the ion composition (abundance) of
 736 a planetary ionosphere. In the Earth's ionosphere, the dissociative recombination of the
 737 molecular O₂⁺, N₂⁺, and NO⁺ ions is particularly important. While this reaction produces
 738 oxygen atoms and/or nitrogen atoms, some of the produced atoms are meta-stable excited
 739 electronic states, which ultimately lead to the formation of airglow and the aurora, and
 740 some atoms are 'hot atoms' in the ground electronic state with excess kinetic energy that
 741 ultimately contribute to heating of the thermosphere via their collisional relaxation with the
 742 ambient oxygen atoms. In the case of the O₂⁺, the dissociative recombination process can
 743 proceed in as many as 5 branches:





749 $\alpha = 2.0 \times 10^{-7} (300/T_e[\text{K}])^{0.7} \text{ cm}^3 \text{ s}^{-1} (T_e < 1200 \text{ K});$

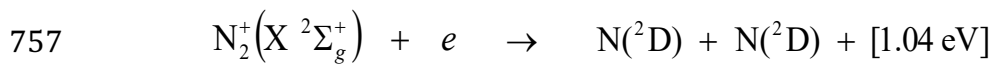
750 $\alpha = 0.74 \times 10^{-7} (1200/T_e[\text{K}])^{0.56} \text{ cm}^3 \text{ s}^{-1} (T_e > 1200 \text{ K})$

751

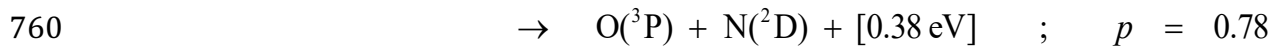
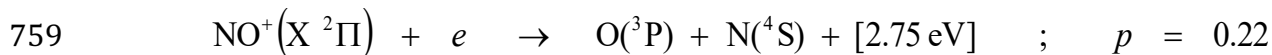
752 where the parameter p denotes the branching ratio, i.e., the fraction going to each branch,
753 the value listed in the square bracket is the excess kinetic energy, and α is the dissociative
754 recombination rate coefficient.

755

756 Similarly, in the case of molecular nitrogen (N_2^+) and nitric oxide (NO^+) ions:



758 $\alpha = 2.2 \times 10^{-7} (300/T_e[\text{K}])^{0.39} \text{ cm}^3 \text{ s}^{-1}$



761 $\alpha = 4.0 \times 10^{-7} (300/T_e[\text{K}])^{0.5} \text{ cm}^3 \text{ s}^{-1}$

762 where the rate coefficient of dissociative recombination α decreases slowly with increasing
763 temperature. In the case of atomic ions, recombination with electrons has to occur via
764 photon emission, and is quite inefficient. For example, the radiation recombination rate
765 coefficient for the radiative recombination of Fe^+ with an electron is $8 \times 10^{-12} \cdot (300/T)^{0.51}$
766 $\text{cm}^3 \text{ s}^{-1}$ (Nahar et al., 1997), which is about 5 orders of magnitude slower than for the
767 typical dissociative recombination reaction of a molecular ion.

768

769 In the auroral ionosphere, the dominant ion production process for atomic O^+ ion is the
770 collisional ionization of neutral atomic oxygen (O) by precipitating auroral electrons,
771 followed by the dissociative ionization of molecular oxygen (O_2). For atomic N^+ , the
772 dominant ion production process is the corresponding dissociative ionization of molecular
773 nitrogen (N_2). Likewise, the dominant ion production process for molecular oxygen and

774 nitrogen ions (O_2^+ and N_2^+) is the electron impact ionization of molecular O_2 and N_2 ,
 775 respectively. Following the treatment of Jones (1974, equation 4.2) the ion production ratio
 776 γ between N^+ and O^+ may be written semi-empirically as (see also Yau et al. 1992):

$$777 \quad (2.1) \quad \gamma = \frac{P_N}{P_O} = \frac{[N_2]}{2.46[O] + 1.46[N_2]}$$

778 where P_N and P_O are the N^+ and O^+ ion production rates, respectively, and $[X]$ denotes the
 779 density of neutral species X .

780
 781 Thus, in the F-region (above ~ 150 km), where $[O_2] \ll [O]$, the production rate ratio γ is
 782 directly proportional to the local neutral molecular nitrogen to atomic oxygen density ratio
 783 $[N_2]/[O]$. During a large magnetic storm and an extended period of auroral substorms, the
 784 neutral N_2 density at auroral latitudes substantially increases (i.e., atmospheric scale
 785 height increases) due to the thermospheric heating. For example, based on the MSIS
 786 model, γ typically increases by a factor of 2–5 from its quiet-time value (A_p index < 5) in
 787 the 300–500 km altitude region during disturbed times ($A_p \sim 100$).

788
 789 All these reactions imply that the combination of (a) auroral electron impact ionization and
 790 dissociative ionization) of molecular N_2 and O_2 and (b) the chemistry of ion-neutral charge-
 791 exchange and of dissociative recombination results in a significant increase in the N_2 to O
 792 density ratio in the F-region and topside auroral ionosphere during the geomagnetic
 793 storms and extended periods of the auroral substorms. Consequently, the molecular ion
 794 density increases in such geomagnetic conditions, resulting in the upflow of the molecular
 795 ions the reduction in overall plasma density in the topside ionosphere.

796

797

798 **2.2.3 Importance of the transport above exobase (above about 500 km)**

799 Except for H^+ and to a lesser extent He^+ , all ions need extra energy to reach the
 800 magnetosphere from the ionosphere. For heavy atomic ions such as O^+ and N^+ , a
 801 substantial portion of the upward moving ions is bounded by gravitation and returns to the
 802 ionosphere (Yamauchi, 2019, and references therein; Dandouras, 2021) before acquiring
 803 sufficient further energy in the main energization region at higher altitudes to overcome
 804 gravity and reach the distant magnetosphere (Delcourt et al., 1993; Yau and André, 1997;
 805 Nilsson et al., 2008; Gronoff et al., 2020).

806

807 Table 2.2 summarizes the escape energy needed for various ions to reach various
 808 altitudes along a vertically oriented geomagnetic field from the ground and from 500 km
 809 altitude (the exobase near solar maximum), respectively, with the latter value given inside
 810 the parenthesis. For example, O_2^+ ions at 500 km altitude flowing along such a field line
 811 needs a minimum of 3.4 eV to reach 2000 km and a minimum of 5.2 eV to reach 3000 km
 812 (actual required energy depends on how much are distributed to the perpendicular
 813 directions), while half the energy is required for the O^+ atomic ions (1.7 eV for reaching
 814 2000 km and 2.6 eV for 3000 km).

815

816 The main energization includes both the electromagnetic wave acceleration (including
 817 mirror acceleration) (Gorney et al., 1982, Yau et al., 1983; Lundin and Guglielmi, 2006)
 818 and electrostatic acceleration (including centrifugal acceleration (Cladis, 1986)), and these
 819 main energization regions vary with time and with locations (latitudes and MLT), with the
 820 lowest altitude about 2000 – 4000 km altitudes.

821

822

823 Table 2.2: Escape velocity (km/s) and energy (eV) for different species and minimum energy to
 824 reach from 500 km altitude (inside parenthesis)

height	escape velocity	O	N ₂	Fe
500 km	10.8 km/s	9.7 eV (0 eV)	17.0 eV (0 eV)	34 eV (0 eV)
1360 km (e-Pop)	10.2 km/s	8.6 eV (1.1 eV)	15.1 eV (1.9 eV)	30 eV (3.8 eV)
1500 km	10.1 km/s	8.5 eV (1.2 eV)	14.8 eV (2.2 eV)	30 eV (4.3 eV)
1700 km (Freja)	9.9 km/s	8.2 eV (1.4 eV)	14.4 eV (2.5 eV)	29 eV (5.0 eV)
2000 km	9.8 km/s	8.0 eV (1.7 eV)	13.9 eV (3.0 eV)	28 eV (6.1 eV)
2500 km	9.5 km/s	7.5 eV (2.2 eV)	13.1 eV (3.8 eV)	26 eV (7.6 eV)
3000 km	9.2 km/s	7.1 eV (2.6 eV)	12.4 eV (4.5 eV)	25 eV (9.0 eV)
4000 km	8.8 km/s	6.4 eV (3.3 eV)	11.2 eV (5.7 eV)	22 eV (11.4 eV)
9000 km	7.2 km/s	4.3 eV (5.4 eV)	7.6 eV (9.4 eV)	15.2 eV (19 eV)
3 R _E	6.5 km/s	3.5 eV (6.2 eV)	6.1 eV (10.9 eV)	12.2 eV (22 eV)
4 R _E	5.6 km/s	2.6 eV (7.1 eV)	4.6 eV (12.4 eV)	9.1 eV (25 eV)
Venus 500 km	9.9 km/s	8.2 eV	14 eV	28 eV
Mars 500 km	4.7 km/s	1.8 eV	3.2 eV	6.4 eV

825

826

827 It is not trivial if the ions can acquire such "pre-energization" because the electric field
828 (both DC and AC) available to energize the ions is normally very small near the exobase.
829 In fact, even upflowing atomic O^+ ions often return back to the ionosphere (Loranc et al.,
830 1991, Yamauchi et al., 2005). There are several mechanisms for the required pre-
831 energization for overcoming gravity above the exobase, but predominantly it is wave-
832 particle interactions. No matter what the mechanism is, the energization is often barely
833 sufficient for a certain ion species to reach the main energization region such that the
834 difference in the m/q by just a factor of two may render the energization insufficient for the
835 heavier mass species to reach the main energization region. If the pre-energization just
836 below 2500 km altitude is only 3–4 eV while the main energization altitudes are different
837 between the dayside and the nightside, the absence of O_2^+ outflow in the nightside can
838 easily be explained. Thus, it is quite possible that such acceleration is sufficient only for
839 atomic ions such as O^+ (and H^+) but not for heavy molecular ions.

840

841 As mentioned above, we expect molecular O_2^+ to reach the main energization region at
842 more equatorward latitudes than atomic O^+ due to the velocity filter effect in the nightside
843 auroral region during geomagnetic storm conditions. Considering that there is less wave
844 activity at lower latitude in the nightside sub-auroral region where O^+ upflow is observed,
845 pre-energization by the wave activity can also be smaller at O_2^+ upflow latitudes than O^+
846 upflow latitudes.

847

848 In addition to the velocity filter effect inside the ionosphere, ionospheric chemistry,
849 ionosphere-thermosphere coupling (see Sects. 2.2.1 and 2.2.2), and upward convection
850 also play roles in determining the distribution of molecular and atomic ions at the topside
851 ionosphere (near and above the exobase). These lead to significant differences for
852 molecular ion distribution between the cusp region and auroral region, possibly allowing
853 molecular ions to leave the ionosphere only from the dayside. Therefore, to understand
854 the magnetospheric molecular ions, we need to know the composition and dynamics of
855 molecular ions at the topside of the ionosphere, too.

856

857 In this respect, it is useful to compare heavy atomic ions (O^+) and heavy molecular ions
858 (N_2^+ , NO^+ , and O_2^+) in terms of total flux only. At high altitude (such as Akebono and Polar
859 altitudes) much above the main energization region, heavy atomic ions (O^+) are found over
860 a wide latitudinal range. Through various route, these upflowing O^+ ions from both the low-

861 latitude ionosphere and high-latitude ionosphere can enter the magnetosphere according
862 to the past observation (e.g., Yamauchi 2019 for review). This is quite different from the
863 heavy molecular ions for which the cusp source dominates in both occurrence and ion flux
864 already at the Akebono altitude (Yau et al., 1993).

865

866 If energy-time profiles can be obtained for molecular ion observations, they give
867 information even on the main energization mechanisms (electrostatic (DC) and
868 electromagnetic (AC)) because they have different mass-per-charge dependences, from
869 no dependence on electrostatic acceleration to mass dependency of ΔE nearly $\sim m^{(-0.5)}$. By
870 knowing the altitude dependence of the flux ratio between different masses and energies,
871 one can estimate the relative importance of different mechanisms at different altitudes,
872 although the feasibility of such measurements is not evident. While the electrostatic (DC)
873 acceleration is normally strong enough to give sufficient energy for all ions (independently
874 of their mass) to reach high-altitude, such DC acceleration does not start below 2000 km
875 altitude, and hence, heavy ions with only a few eV at exobase do not reach the DC
876 acceleration region.

877

878 The detection of heavy ions also gives clues on how low the electromagnetic (AC)
879 acceleration starts. On the other hand, AC acceleration might start from much lower
880 altitudes during magnetic storms or other conditions, but details are unknown. If AC
881 acceleration is strong enough and starts at low enough altitudes, even metallic ions may
882 reach the DC acceleration region. Therefore, quantification of heavy ions in the
883 magnetosphere provides information on the strength and altitudes of the AC acceleration.

884

885

886 **2.2.4 How much change of atmospheric composition (e.g., N₂/O₂ ratio) on geological** 887 **scales is caused by the ion escape?**

888

889 Evolution of the Earth's atmosphere is another science theme that benefits from our
890 knowledge of the magnetospheric molecular ions (N₂⁺, NO⁺, and O₂⁺), in addition to atomic
891 heavy ions N⁺ and O⁺ (e.g., Yamauchi, 2019). The present atmospheric composition is
892 quite different from that of the other planets after 4.5 billion years of evolution (e.g., Mars
893 and Venus for which the initial atmospheric composition is believed to be similar to that of
894 the Earth, Lammer et al. (2008)). The difference in the atmospheric evolution must have

895 influenced the habitability of these three planets: very limited habitability in best case for
896 Mars (underground where water exists) and Venus (cloud layer where temperature is in
897 the habitable range) compared to the full habitability for all known forms of life on the
898 Earth.

899

900 From the viewpoint of evolution of life, a very small change of the atmospheric composition
901 is important. A change of only a few percent in the atmospheric O₂/N₂ ratio or in the water
902 pH, or a change of a few degrees in the atmospheric and water temperature may
903 significantly affect the biochemical reactions and hence metabolism and photosynthesis
904 (e.g., Loesche, 1969; Hill, 1976; Servaites, 1977; Ku et al., 1977; Harrison, 2010). A
905 change of a few percent in the atmospheric N/O ratio corresponds to a change of about
906 10% in the nitrogen inventory in the present-day biosphere ($(4-5) \times 10^{18}$ kg). This amount of
907 loss is achieved by 600 million years ($=2 \times 10^{16}$ sec) of average nitrogen loss rate of 10^{27} s^{-1}
908 (about 20 kg/s). The same level of change (15% fluctuation in O₂ content) has actually
909 occurred during a duration of 100 million years in the past according to the geological
910 record (Berner, 2006). These durations and amount are short enough compared to the
911 history of life and large enough in quantity, respectively, to affect bacteria through change
912 in the N/O ratio in the atmosphere.

913

914 In addition, the simultaneous presence of significant amounts of N₂ and O₂ in an
915 atmosphere is chemically incompatible over geological timescales, and hence, the present
916 N₂/O₂ ratio must have resulted from the biological activity (Lammer et al., 2019; Stüeken et
917 al., 2020; Dandouras et al., 2020), constituting a bio-signature.

918

919 The question is then what causes such changes and fluctuations of the atmospheric
920 composition. There are six main channels that determine the evolution of the atmosphere:
921 (a) net escape to space after removing the return flow; (b) net influx from space (e.g.,
922 meteors); (c) biospheric reactions (e.g., O₂ from photosynthesis); (d) sub-surface sink
923 through ocean bottom; (e) emission from sub-surface through both non-organic (e.g.,
924 volcanic) and organic activities (bacterial denitrification) (Berner, 1999; Canfield, 2005;
925 Barabash et al., 2007; Johnson and Goldblatt, 2015); and (f) geochemistry (e.g. CO₂
926 chemical capture through limestone formation) (Stüeken et al., 2020).

927

928 Among those, the net contribution of the biosphere (c) and from the Earth's interior (d and
929 e) diminishes once the photosynthesis and mantle convection are stabilized because
930 these contributions mainly recycle elements rather than causing net changes. Also, the net
931 influx of biological elements (N, O, C) from space (b) is much smaller than that of escape
932 to the space. On the other hand, (a) the escape to space causes net changes, and
933 therefore its relative importance compared to the sub-surface migration is large in the
934 geological time scale. The question is: how much?

935

936 The aforementioned escape rate to space of 10^{27} s^{-1} is not unrealistic during the ancient
937 time of about 4 billion years ago, because the composition and amount (flux) should
938 depend strongly on the solar UV, solar wind, and geomagnetic activity conditions, all of
939 which are known to be much higher in the ancient than present (Yamauchi, 2019;
940 references therein). This means that we can assume frequent Kp ~ 10 at that time (Krauss
941 et al., 2012, Slapak et al., 2017). Due to increased frequency of high Kp activity, Kp ~ 10
942 can be used, the increased escape can be assumed almost continuously, rather than a
943 summation of short-lived events of "massive escape" for the present days. Assuming
944 escape rate decreasing linearly with time, Slapak et al. (2017) estimated the total loss of
945 heavy atomic ions (O^+ and N^+) over 4 billion years as about $5 \times 10^{17} \text{ kg}$, which corresponds
946 to 40% of today's total oxygen mass in the atmosphere.

947

948 In such a condition, i.e., during an enhanced outflow rate, energization of the ionosphere is
949 also enhanced, allowing even molecular ions (N_2^+ , NO^+ , and O_2^+) to gain sufficient initial
950 velocity to enter the outflow process (few-ten eV) within their relatively short dissociative
951 recombination lifetime. Therefore, during "massive escape" events, flux enhancement of
952 the ion outflow becomes more drastic for molecular ions than for atomic ions (and within
953 atomic ions, N^+ outflow is more enhanced than O^+ outflow). These factors cause higher
954 N/O ratio of the atmospheric loss during enhanced outflow rate, and hence in the ancient
955 time in the geological scale.

956

957 The high N/O ratio of escaping elements is consistent with what has been observed in the
958 very sparse observations that simultaneously detected the molecular ions (Yau et al. 1993,
959 Hamilton et al., 1988). However, no quantitative value for N/O ratio or even
960 molecular/atomic ratio for ion outflow flux (or upflow flux at low altitude) is available due to
961 a lack of proper instruments in the magnetospheric missions covering a wide variety of

962 escape routes and respective energy ranges. From the detection capability viewpoint,
 963 quantitative separation of N^+ and O^+ requires much higher $m/\Delta m$ capability than separating
 964 molecular ions (N_2^+ , NO^+ , or O_2^+) from atomic ions (N^+ or O^+), but capable instruments
 965 have already flown on board planetary missions such as Cassini, Kaguya, MAVEN, Bepi-
 966 Columbo, and JUICE.

967

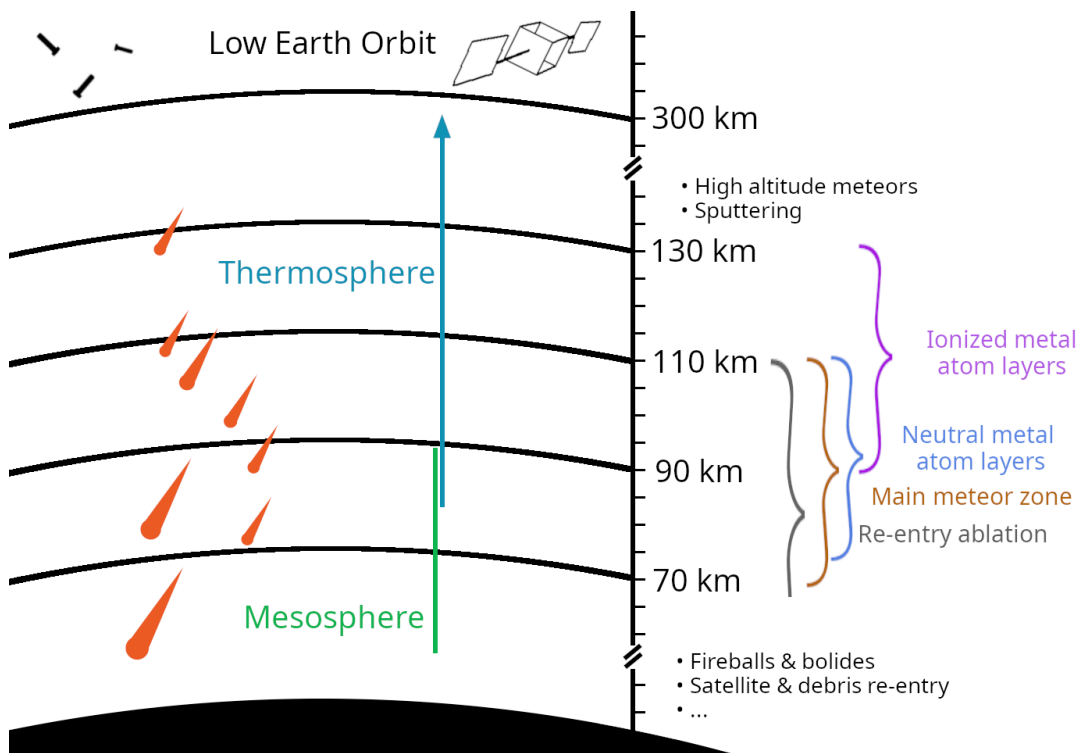
968

969 2.3 Importance and present knowledge of the mesospheric sources (metallic ions)

970

971 Figure 2.1 demonstrates the existence of accelerated (> 100 keV) low charge-state
 972 metallic ions (such as Fe^+) in the magnetosphere, although their flux is much lower than
 973 the flux of the solar wind metallic ions entering the magnetosphere. This also indicates that
 974 some ions with $m/q \approx 30$ can also be Si^+ . The low charge-state indicates that they must
 975 come from either from the Moon (see Sect 2.4) or the atmosphere. Although metallic
 976 species are a very minor component of the atmosphere, there are several layers of
 977 metallic species in the mesosphere and lower thermosphere, mainly from the ablation of
 978 cosmic dust entering the Earth's atmosphere, as illustrated in Figure 2.5.

979



980

981 Fig2.5: Illustration of the ablation of material entering the atmosphere, showing relevant

982 altitude ranges.

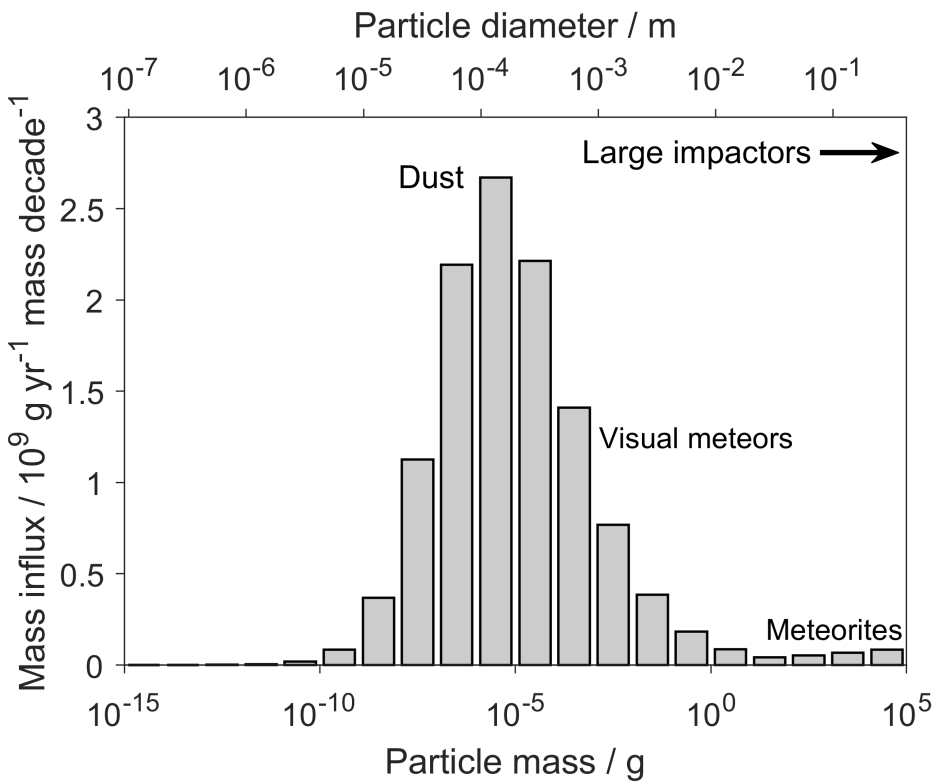
983

984

985 **2.3.1 Meteoroids and the metal layers**

986 Every day the Earth's atmosphere is bombarded by billions of dust-sized particles and
 987 larger pieces of material from space. The primary sources of these cosmic dust particles
 988 are the sublimation of volatile species in comets as they are heated by the Sun, which
 989 release dust particles that are then ejected by drag forces, and collisions between
 990 asteroids in the main asteroid belt between Mars and Jupiter (Plane et al., 2018b). These
 991 particles are collectively termed meteoroids. Because the particles range in size by more
 992 than 12 orders of magnitude as shown in Figure 2.6 (Murad and Williams), estimating the
 993 total input of cosmic dust into the atmosphere is very challenging, with estimates ranging
 994 from roughly 5 – 300 t d⁻¹ (tonnes per day) (Plane et al., 2012).

995



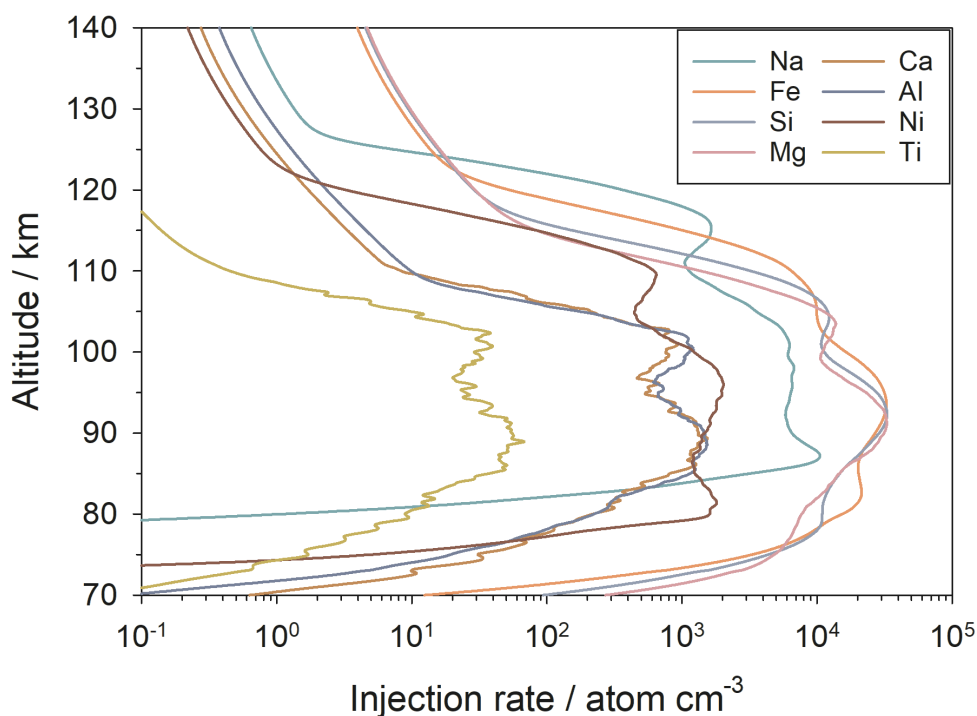
996

997 Figure 2.6. The annual meteoroid mass influx (expressed per decade of mass) plotted
 998 against particle mass, adapted from Plane et al. (2018b) and Schulz and Glassmeier
 999 (2021).

1000

1001

1002 The most likely value currently is thought to be around 30 t d⁻¹, based on measurements of
 1003 the vertical fluxes of Na and Fe atoms which ablate from dust in the lower thermosphere
 1004 and upper mesosphere, and the accumulation of cosmic spherules (dust particles that melt
 1005 but do not completely evaporate) at the South Pole (Carrillo-Sánchez et al., 2020). This
 1006 input rate is corroborated by the optical extinction of meteoric smoke particles (which form
 1007 from the polymerisation of metallic compounds produced from ablated metal atoms) in the
 1008 lower mesosphere (Hervig et al., 2021), and the accumulation of unmelted
 1009 micrometeorites in a large collection from Concordia in Antarctica (Rojas et al., 2021) as
 1010 well as earlier estimates considering their respective biases (Schulz and Glassmeier,
 1011 2021, and references therein).
 1012



1013
 1014 Figure 2.7. Global average ablation rates as a function of altitude for the main metal
 1015 constituents in cosmic dust (Carrillo-Sánchez et al., 2020).
 1016

1017 As meteoroids enter the atmosphere, they undergo heating by inelastic collisions with air
 1018 molecules. If they reach the melting point (~1800 K), then ablation (i.e., evaporation) of the
 1019 constituents becomes rapid. Initially the relatively volatile elements (Na and K) ablate,
 1020 followed by the main constituents (Fe, Mg and Si) around 2000 K. Finally, the refractory
 1021 elements (Ca, Al and Ti) ablate if the particle reaches a temperature over 2400 K (Vondrak

1022 et al., 2008). Figure 2.7 shows the ablation rates of the individual elements as a function of
1023 altitude, illustrating that most ablation occurs between 70 and 110 km (Carrillo-Sánchez et
1024 al., 2020). This injection is the source of the layers of neutral metal atoms that occur
1025 globally between about 75 and 110 km, and the layers of ionized metal atoms between
1026 about 90 and 130 km (Plane, 2003; Plane et al., 2015a)

1027

1028 Metallic atoms have comparatively low ionization energies. Since they are initially
1029 travelling at the same speed as their parent meteoroid, they can undergo collisional
1030 ionization with air molecules. The meteoroid energy velocity ranges from 11 km s⁻¹ to 72
1031 km s⁻¹, and the ionization probability strongly depends on the velocity (see Figure 3 in
1032 Janches et al., 2017). The resulting dense plasma, together with optical emissions from
1033 atoms and molecules in excited electronic states, is termed the meteor, which moves
1034 together with the ablating meteoroid (Ceplecha et al. 1998). If the meteoroid survives the
1035 ablation process and reaches the ground, it is termed a meteorite. Note that the analysis
1036 of meteorites and observations of meteor spectra provide conclusive evidence that the
1037 meteoroid population contains metallic elements roughly in same abundance as the
1038 photosphere (Asplund et al., 2009; Lodders et al., 2009, Kero et al., 2019).

1039

1040 There have also been rare observations of the meteor phenomena at altitudes up to 200
1041 km. For optical observations these events have been mainly explained by sputtering (non-
1042 thermal ablation) (Popova et al., 2007). With sufficient meteoroid size and velocity,
1043 sputtering can create enough photons to be detected by optical systems (Koten et al.,
1044 2006). The majority of published optical observations of high altitude meteors originate
1045 from the Leonid meteor shower due to its very high velocity (about 70 km s⁻¹). These
1046 events are also characterized by the sputtering-dominated part of the meteor event with a
1047 brightness that is several orders of magnitude fainter than the thermal ablation part,
1048 indicating that a much smaller part of the meteoroid mass is lost at these altitudes
1049 (Vondrak et al., 2008). High altitude meteors have also been observed with radars.
1050 However, the very high-altitude cases (>150 km) are still an open question due to possible
1051 ambiguities in the observation techniques (Brosch et al. 2013; Vierinen et al. 2014; Gao
1052 and Mathews 2015). Radar observations of high-altitude meteors up to 142 km have been
1053 validated with respect to such ambiguities (Kastinen and Kero 2022) and may be
1054 explained by thermal ablation, particle disruption, and/or the pyrolysis of refractory
1055 organics within the dust particles (Bones et al., 2022).

1056

1057 To determine the altitude and element distribution of mass deposition in the atmosphere,
1058 the meteoroid atmospheric entry needs to be modelled. For an overview of modelling the
1059 atmospheric entry of meteoroids (Ryabova et al., 2019, and references therein). These
1060 models have been widely used for determining the physical properties of meteoroids from
1061 observations (e.g., Gritsevich 2009; Campbell-Brown et al., 2013), and for estimating the
1062 injection flux of metals into planetary atmospheres as input when modelling the
1063 atmospheric chemistry and dynamics in the mesosphere and lower thermosphere (Plane
1064 et al., 2015a). In recent years, chemical ablation models have been tested using different
1065 types of laboratory meteoric ablation simulators (Gomez Martic et al., 2017; Thomas et al.,
1066 2017) to constrain chemical ablation models, such as the CABMOD model which now
1067 includes separate metal silicate and Fe-Ni-S phases (Bons et al., 2019).

1068

1069 In Hulfeld et al., (2021), the break-up of dustball meteoroids was simulated using fluid
1070 dynamics simulations of the meteoroid's atmospheric entry flow, including both thermal
1071 and mechanical break-up mechanisms. A Draconid meteoroid was simulated starting with
1072 compression by the aerodynamic forces to approximately half its size at the beginning of
1073 the simulation (200 km altitude), and then mechanically disintegrated at 120 km altitude
1074 due to aerodynamic-induced meteoroid rotation. In contrast, camera and radar
1075 observations of Draconids show a lower break-up altitude of ~ 100-110 km (Borovička et
1076 al. 2007, 2014; Kero et al. 2012). These studies indicate that there may be an additional
1077 influx of meteoroid material that disperses (=ablates) at higher altitudes than regular
1078 thermal ablation of a single solid body allows.

1079

1080

1081 **2.3.2: Observations of metallic ions in the thermosphere/ionosphere**

1082 Metallic ions in the lower thermosphere (lower ionosphere) have long been detected and
1083 observed by both in-situ observation (rockets and satellites) and remote observation
1084 (ground and satellites), although the spatial and temporal resolution at higher altitude is
1085 insufficient to reveal the vertical transport.

1086

1087 **(a) Spaceborne observations**

1088 For in-situ sampling, the mass spectrometer on board Atmosphere Explorer C (AE-C)
1089 satellite detected patches of Fe⁺ between 220 and 320 km that seemed to be associated

1090 with regions of upward plasma transport (Grebowsky and Brinton, 1978). Moreover, the
1091 retarding potential analyser (RPA) on board OGO-6 satellite occasionally observed Fe⁺ at
1092 much higher altitudes between 600 and 950 km (Hanson and Sanatani, 1971; Hanson et
1093 al., 1972). Measurements of metallic ions up to around 130 km have been made using ion
1094 mass spectrometers on sub-orbital rockets, mostly at mid- and high-latitude locations. This
1095 type of experiment provides vertical profiles with typically 2 km height resolution of all ions
1096 with masses below ~100 amu and concentrations larger than ~10 cm⁻³, but not at middle
1097 and upper ionosphere. These measurements were mostly made in the 1970s and 1980s
1098 (for review, Kopp, 1997; and Grebowsky and Aikin, 2002).

1099
1100 Metallic species such as Mg⁺ and Fe⁺ have also been observed by resonant scattering of
1101 sunlight, for example by the Space Shuttle (Gardner et al., 1995; 1998). An optical
1102 spectrometer, ARGOS/ISAAC, detected clear Fe⁺ emission in the 100-340 km altitude
1103 region (Dymond et al., 2003). In the last 15 years, spaceborne limb-scanning optical
1104 spectrometers have been used to determine the vertical profiles of metal atoms and ions:
1105 the Optical Spectrograph and Infra-Red Imager System (OSIRIS) spectrometer on the
1106 ODIN satellite for Na (Hedin and Gumbel, 2011) and K (Dawkins et al., 2014); and the
1107 Scanning Imaging Absorption Spectrometer for Atmospheric Chartography (SCIAMACHY)
1108 spectrometer for Mg and Mg⁺ (Langowski et al., 2015) and the Global Ozone
1109 Measurement by Occultation of Stars (GOMOS) spectrometer for Na (Fussen et al., 2010),
1110 both on the Envisat satellite.

1111

1112 **(b) Resonance lidars**

1113 The first measurements of metal atom densities (Na, K, Fe and Ca⁺) in the upper
1114 mesosphere were made in the 1950s using ground-based twilight photometry, where
1115 resonance fluorescence from spectroscopic transitions of metal atoms excited by solar
1116 radiation was measured during twilight (Hunten, 1967). These measurements were
1117 superseded in the 1970s when the development of tunable lasers led to the resonance
1118 lidar technique (Plane, 1991). Lidar has been used to observe Na, K, Li, Ca, Ca⁺ and Fe
1119 (Plane, 1991), and recently Ni (Gerding et al., 2019). Most of these observations have
1120 focused on the main metal layers between 75 and 110 km.

1121

1122 One important development in the past 15 years has been the use of high performance
1123 lidars to extend observations into the thermosphere of Fe (Schneider et al., 2015) and Na

1124 (Liu et al., 2016) above 150 km, K and Ca up to 130 km (Höffner and Friedman, 2004,
 1125 2005; Friedman et al., 2013), and Ca⁺ up to 180 km (Raizada et al., 2020). Even the
 1126 diurnal observations became possible (Plane, 2003). Thus, it has long known that these
 1127 metallic species exist at detectable levels in the lower thermosphere/ionosphere.

1128

1129

1130 2.3.3 Transport to the exobase (old sec 5.3.2)

1131 In the thermosphere, metals exist almost entirely as metallic ions after the efficient
 1132 ionization either by charge exchange with ambient NO⁺ and O₂⁺ ions, or by photoionization
 1133 (Plane et al., 2015a). Unlike the molecular ions such as NO⁺ which undergo rapid
 1134 dissociative recombination with electrons, metallic ions can only undergo dielectronic or
 1135 radiative recombination with electrons, a process about 10⁶ times longer lifetime (Plane et
 1136 al., 2015a). Thus, the metallic ions have lifetimes of days in the thermosphere.

1137

1138 Table 2.3 Metallic elements and their mass

ions	mass	molecular ions with same mass	space debris
Na ⁺	m=23		
Mg ⁺	m=24		
Al ⁺	m=27	HCN ⁺	yes
Si ⁺	m=28	N ₂ ⁺ , CO ⁺	yes
P ⁺	m=31	¹⁵ N ¹⁶ O ⁺ or ¹⁴ N ¹⁷ O ⁺	
S ⁺	m=32	O ₂ ⁺	
K ⁺	m=39		
Ca ⁺	m=40		
SiO ⁺	m=44	CO ₂ ⁺	
Ti ⁺	m=48		yes
Fe ⁺	m=56		yes
Ni ⁺	m=59		yes
Cu ⁺	m=64		yes
Ge ⁺	m=73		yes

1139

1140

1141 Furthermore, some metallic ions like Na⁺ (m=22), Mg⁺ (m=24), Al⁺ (m=27), Si⁺ (m=28), P⁺
 1142 (m=31), and S⁺ (m=32) have similar or even larger m/q compared to heavy molecular ions,
 1143 allowing them lifted upward longer distances than heavy molecular ions within the lifetime.
 1144 Such long upward distance even applies to the other relatively abundant heavy metallic
 1145 ions, such as K⁺ (m=39), Ca⁺ (m=40), SiO⁺ (m=44), Fe⁺ (m=56), Ni⁺ (m=59), Cu⁺ (m=64),
 1146 because they have masses that are within a factor of two of the heavy molecular ions

1147 listed in Table 2.3 (here, we note that mass 44 in the magnetosphere is not necessarily
1148 CO_2^+ , but could also be SiO^+ of lunar origin).

1149

1150 Since molecular ions must be transported from the lower ionosphere to reach the exobase
1151 within the dissociation timescale, the same "strong" upward ion convection in the
1152 ionosphere may also transfer metallic ions against the gravity (particular those lighter than
1153 O_2^+) from the lower ionosphere. Inversely, the detection of the heavy molecular ions
1154 ($m/q \approx 30$) above the ionosphere by e-POP suggests that metallic ions may also access the
1155 topside ionosphere (exobase). If one can measure the ratio of metallic ions and molecular
1156 ions of similar masses in the ion outflow above the ionosphere, the variation of this ratio
1157 indicates change in the convection and/or change in the chemical reaction.

1158

1159 Unfortunately, no dedicated instrument or mission exists in a terrestrial orbit to monitor
1160 such ions leaving the ionosphere or arriving at the topside ionosphere from lower altitudes.
1161 Meanwhile, modeling efforts have advanced to include the metallic ions and atoms, as
1162 described in Sect 4.3.3.

1163

1164

1165 **2.3.4 Possible human-made contribution by atmospheric re-entry of space debris**

1166 Out-of-service satellites, rocket bodies, and subsequent fragmented parts, together
1167 constitute space debris. Their re-entering into Earth's atmosphere causes the same
1168 ablation process as the entry of meteoroids although with much smaller entry velocities (\sim
1169 8 km/s), shallower entry angles and different composition, resulting in the deposition of
1170 metallic atoms and ions into the upper atmosphere (Schulz and Glassmeier, 2021).

1171 Returning spacecraft with astronauts and goods may experience the same process, but
1172 the degree of ablation is much smaller than that of space debris for which re-entry is
1173 designed to result in complete ablation, and therefore this is included in the category of
1174 "ablation of space debris" or a wider terminology of "ablation of space waste".

1175

1176 Currently, the mass injected into the atmosphere by re-entering space debris is only a
1177 fraction (about 3 % in 2019) of what is injected by meteoroids (Schulz and Glassmeier,
1178 2021). However, metallic species are present in much higher fraction in space debris than
1179 in meteoroids, resulting in the atmospheric re-entry flux of some metallic species (mainly

1180 Al and Li, but also Ni, Cu, Ti, and Ge) from space debris surpassing the entering flux of the
1181 same species from meteoroids. For these species, the annual mass input to the whole
1182 atmosphere will exceed or has already exceeded the natural input considering the strong
1183 increase in launch activity every year. The details of such future increase and its
1184 implications are discussed in section 4.3.2. All this poses the question how much the input
1185 of re-entering space debris contaminates the natural origin metallic atoms and ions in the
1186 mesospheric metal layers. To answer the question, knowledge about the ablation
1187 characteristics of entering space debris and the subsequent chemical processes are vital.
1188

1189 For re-entering space debris, ablation starts as high as 110 km (Fritsche et al. 2000,
1190 Klinkrad 2005, Rafano Carná and Bevilacqua 2018) which is 20 km lower than the
1191 meteoroid cases. For larger spacecraft (few 100 kg or more), the main part of mass loss
1192 takes place at altitudes below 80 km depending on the spacecraft velocity, entry angle,
1193 mass, and composition (Reynolds et al. 2001, Lips et al. 2005, Battie et al. 2013,
1194 Buttsworth et al. 2013, Jenniskens et al. 2016 and Park et al. 2021). For example, debris
1195 on a very elliptic orbit (high-apogee) enters the atmosphere with higher velocity and angle
1196 than low-Earth orbit (LEO) debris. Note that Buttsworth et al. (2013) and Jenniskens et al.
1197 (2016) considered non-typical re-entries (very high apogee orbits, thus a very high entry
1198 velocity and angle compared to LEO spacecraft), allowing the spacecraft survive to lower
1199 altitudes for long-lasting ablation.

1200
1201 Metallic atoms that ablate below 80 km will quickly be oxidized to metal oxides, hydroxides
1202 and carbonates (Grebowsky et al., 2017). For instance, an Fe atom that ablates at around
1203 64 km will be oxidized to FeO by O₃ in about 2 s (Grebowsky et al., 2017), and Al will be
1204 oxidized by reaction with O₂ much more rapidly (Plane et al., 2021). Oxidized metallic
1205 species that form below 80 km as a result of the ablation of space debris will rapidly
1206 polymerize with themselves or the background population of nanometer-sized meteoric
1207 smoke particles that are produced from meteoric ablation (Plane et al., 2021). These tiny
1208 particles will then be transported down to the Earth's surface by the residual atmospheric
1209 circulation. An increase in the number and size of these particles as a result of space
1210 debris may have some effect on the stratospheric ozone layer (e.g. freezing polar
1211 stratospheric clouds droplets (James et al., 2018) or removing the main chlorine reservoir
1212 HCl (Plane, 2003)). However, the particles are very unlikely to be transported above 90
1213 km, such that they provide a source of thermospheric metallic ions after decomposing.

1214

1215 In contrast, for metals that ablate from space debris above 80 km, the high background
1216 concentrations of atomic O and H in the upper mesosphere (Plane, 200) will maintain a
1217 high level of metal atoms (or AlO in the case of Al) because they reduce metal oxides and
1218 other compounds. These metals may then be transported to the lower thermosphere,
1219 analogously to the transport of ions from the natural metal layers, and ionized.

1220

1221

1222

1223 **2.4 Importance of the Moon as source and present knowledge (sputtering and** 1224 **pickup process)**

1225

1226 The Moon is a source of heavy ions. These ions are generated on the lunar surface either
1227 directly sputtered from the lunar surface by impact of solar wind plasma or magnetospheric
1228 plasma (Yokota et al., 2009, Wieser et al., 2010), or by the photoionization (solar UV and
1229 EUV) of the neutral exosphere (Stern, 1999). These exospheric neutrals are originally
1230 generated by (1) micro-meteorite impact vaporization, (2) solar photon stimulated
1231 desorption, (3) sputtering by the solar wind and magnetospheric ions, and (4) thermal
1232 desorption (Colaprete et al. 2016; Wurz et al. 2022).

1233

1234 Since these Moon-origin ions are typically high mass but singly charged, m/q values are
1235 much higher than those of the solar wind ions (all species have similar m/q within a factor
1236 of 2). Therefore, the gyroradius of the Moon-origin ions after gaining the solar wind speed
1237 due to the pick-up process is quite different from those of the solar wind. This uniqueness
1238 of the lunar metallic species makes even a tiny amount of the ion as a tracer to give extra
1239 information in the ion dynamics at the magnetospheric boundary.

1240

1241

1242 **2.4.1 Formation of the exosphere and ion pickup**

1243 The lunar exosphere, although very thin according to Apollo observations (Stern, 1999),
1244 has been observed by Lunar Atmosphere and Dust Environment Explorer (LADEE)
1245 mission (Mahaffy et al., 2015; Halekas, et al., 2015). The column densities of alkali
1246 elements (K and Na) on the Moon observed by LADEE increased during the meteor
1247 shower events during its mission from November 2013 to April 2014 (Colaprete et al.

1248 2016). For the Leonid meteor shower (November) and Geminid meteor shower
1249 (December), response was sharper for K than Na, whereas for Quadrantid meteor shower
1250 (January) the response was small and nearly the same for both elements. These meteor
1251 showers are more important than CIRs or CMEs for the production of the exospheric
1252 neutrals (Colaprete et al. 2016).

1253

1254 As for the composition, Neutral Mass Spectrometer (NMS) on board LADEE has reported
1255 observations of lunar exospheric ions (at low energies < 25 eV) in the lunar orbit when the
1256 Moon was in the solar wind. Using the dedicated ion mode (Mahaffy et al., 2015), ions of
1257 expected species of the lunar exosphere were recorded at masses 2 (H_2^+), 4 (He^+), 16
1258 (O^+), 20 (Ne^+), 23 (Na^+), 39 (K^+), and 40 (Ar^+) amu, but also of unexpected ions including
1259 $^{12}\text{C}^+$, $^{14}\text{N}^+$ and at mass 28, which could be Si^+ , N_2^+ or CO^+ (the presence of ^{12}C (Halekas et
1260 al., 2015) suggests that it is most likely CO^+). These observed ions originate from the
1261 exosphere, rather than directly from the surface (Mahaffy et al., 2015).

1262

1263 Lunar Dust Instrument (LDEX) on board LADEE showed that the electric current, which is
1264 most likely monitoring the lunar pickup ions, is linearly correlated with the solar wind flux
1265 (Poppe et al., 2016b, Fig. 3). The reported ion fluxes are best fit by total exospheric ion
1266 production rates of about $6 \times 10^3 \text{ m}^{-3} \text{ s}^{-1}$. Since LDEX does not have a means to identify
1267 the mass of the recorded ions, the ion composition was inferred based on modelling with
1268 dominant contributions from Al^+ , CO^+ , and Ar^+ .

1269

1270

1271 **2.4.2 Sputtering from the lunar surface**

1272 The sputtering from the lunar surface is theoretically expected (Yokota and Saito 2005;
1273 Futaana et al., 2006; Wurz et al., 2007) and actually observed by Kaguya (SELENE) and
1274 Chandrayaan-1 lunar orbiters. Kaguya Ion Mass Analyzer (IMA) detected the sputtered
1275 ions of many species together with the ionized exospheric neutrals and the reflected solar
1276 wind ions (Yokota et al., 2009; Tanaka et al., 2009). Chandrayaan-1 Sub-keV Atom
1277 Reflecting Analyser (SARA) detected sputtered neutral hydrogen and heavy atoms of
1278 oxygen mass group (Wieser et al., 2010; Vorburger et al. 2014).

1279

1280 The sputter yields from general surfaces are energy dependent, and approaches zero for
1281 lower energies, because the energy deposited in the surface by the impacting ion is not

1282 sufficient to overcome the binding energy of atoms at the surface. The sputter yield also
1283 approaches zero for very high energies because high-energy ions penetrate deeper into
1284 the solid without depositing significant energy at and near the surface to cause the release
1285 of particles. The maximum yield is expected at an energy around 1 keV/nuc of the
1286 impacting ions for the lunar case (Wurz, 2012; Wurz et al. 2022), which compatible with
1287 ions with the solar wind velocity (for all species) and with the thermal magnetospheric
1288 plasma (light ions).

1289

1290 This means that we expect effective sputtering from the lunar surface when the Moon is
1291 exposed to the solar wind. Particularly, we expect drastic increases in the surface
1292 sputtering by CMEs (Leblanc et al., 2022) because a CME is accompanied by drastic flux
1293 enhancements of the heavy ion component: the degree of flux increase is much greater
1294 than that for H^+ (Wurz et al., 2001, 2003; Wimmer-Schweingruber et al., 2006). The
1295 effective sputtering is even expected in the magnetospheric plasma lobe or
1296 magnetosheath. On the other hand, contribution by EUV (ionization of exospheric heavy
1297 atoms by EUV) should not cause such drastic change compared to the sputtering
1298 contribution because the solar flare that is very short-lived.

1299

1300 Elphic et al. (1991) conducted a laboratory study of the ion emission caused by ion impact
1301 on materials (sputtering) with solar wind-like ion and material analogue to the lunar
1302 surface. Using H^+ and He^{++} primary ions, they found that these ions produce significant
1303 fluxes of sputtered ions (so-called secondary ions) of lunar surface material, including Na^+ ,
1304 Mg^+ , Al^+ , Si^+ , K^+ , Ca^+ , Ti^+ , Mn^+ , and Fe^+ , although H^+ and He^{++} are not efficient sputterers.
1305 The predicted secondary ion fluxes from the lunar surfaces is between ~ 10 and 10^4 ions
1306 $cm^{-2} s^{-1}$, depending on the species. Thus, the range of relative ion yields covers four digits
1307 of variation depending on sputtered ion species. Similar studies were performed on Apollo
1308 soils (soil number 10084 and soil number 62231) and on a synthetic Corning glass lunar
1309 simulant (Dukes and Baragiola, 2015). X-ray photoelectron spectroscopy was correlated
1310 with the spectra of secondary ions ejected from these soils by 4 keV He ions. The ejected
1311 secondary ion species from the Apollo soils by 4 keV He include the atomic ions: Na^+ ,
1312 Mg^+ , Al^+ , Si^+ , Ca^+ , Ca^{++} , Ti^+ , Fe^+ , and molecular the ions: NaO^+ , MgO^+ and SiO^+ .

1313

1314 Yokota and Saito (2005) modelled the ion production near the Moon, at 100 km above the
1315 surface, including photoionization of the lunar exospheric atoms, photon-stimulated ion

1316 desorption, and ion sputtering. They proposed that an intense flux of picked-up lunar ions
 1317 ($10^4 \text{ cm}^{-2} \text{ s}$) exists at an altitude of 100 km, for nearly a quarter of the orbit, with the main
 1318 contributions from Na^+ , Mg^+ , Al^+ , Si^+ , K^+ , Ca^+ , Ti^+ , Mn^+ , and Fe^+ ions. In the other model
 1319 by Sarantos et al. (2012), ion species of Ti^+ , Fe^+ , Mg^+ , and especially Ca^+ are mainly
 1320 ejected from the surface, and ionization of the exospheric constituents the other species,
 1321 leading to the estimated fluxes that significantly exceed the ion production rate at the
 1322 surface. These sputtering yields also depend on the lunar geographical areas (Futaana et
 1323 al., 2006) as confirmed by Chandrayaan-1 (Wieser et al., 2010).

1324

1325

1326 **2.4.3 Amount and composition of ions leaving the Moon: Kaguya observation**

1327 Kaguya/IMA made the first in-situ detection of the heavy ions originating from the lunar
 1328 surface and exosphere in a polar orbit with an altitude of 100 km, 50 km, and in an
 1329 elliptical orbit with perigee altitude as low as 10 km (Saito et al., 2010). Kaguya/IMA is
 1330 capable of detecting Moon-origin ions (both sputtering ions directly from the surface and
 1331 pickup ions in the terminator region) for energy up to 12 keV/q. In their observation at 100
 1332 km altitude, Kaguya/IMA detected heavy ions including C^+ , O^+ , Na^+ , K^+ , and Ar^+ originating
 1333 from the Moon surface/exosphere (Yokota et al., 2009; Lee et al., 2024) when the Moon
 1334 was in the solar wind.

1335

1336 The Kaguya/IMA observation confirms that the sputtered ions have energies of about a
 1337 few hundred eV in most cases, in agreement with the above expectation (Yokota et al.,
 1338 2009, 2020; Tanaka et al., 2009). Figure 2.8a shows an example of such Kaguya
 1339 observation. However, when a CIR (Corotating Interaction Region) passed the Moon
 1340 carrying enhanced IMF and solar wind speed (which causes also an enhanced convection
 1341 electric field), the energy of the sputtered heavy ions can become even higher than the
 1342 incident solar wind proton energy, as shown in Figure 2.8b. More importantly, the ion flux
 1343 of heavy ions drastically increased during the CIR passage, as shown in Figure 2.9 (same
 1344 events as Figure 2.8). Note that the CIR contains H^+ , He^{++} , He^+ , C^+ , O^+ , Na^+/Mg^+ , Al^+/Si^+ ,
 1345 P^+/S^+ , K^+/Ar^+ , Mn^+/Fe^+ . Existence of the high-energy low charge-state metallic heavy ions
 1346 associated with CIRs indicates that the contribution of the solar wind sputtering becomes
 1347 important when the solar wind pressure is high.

1348

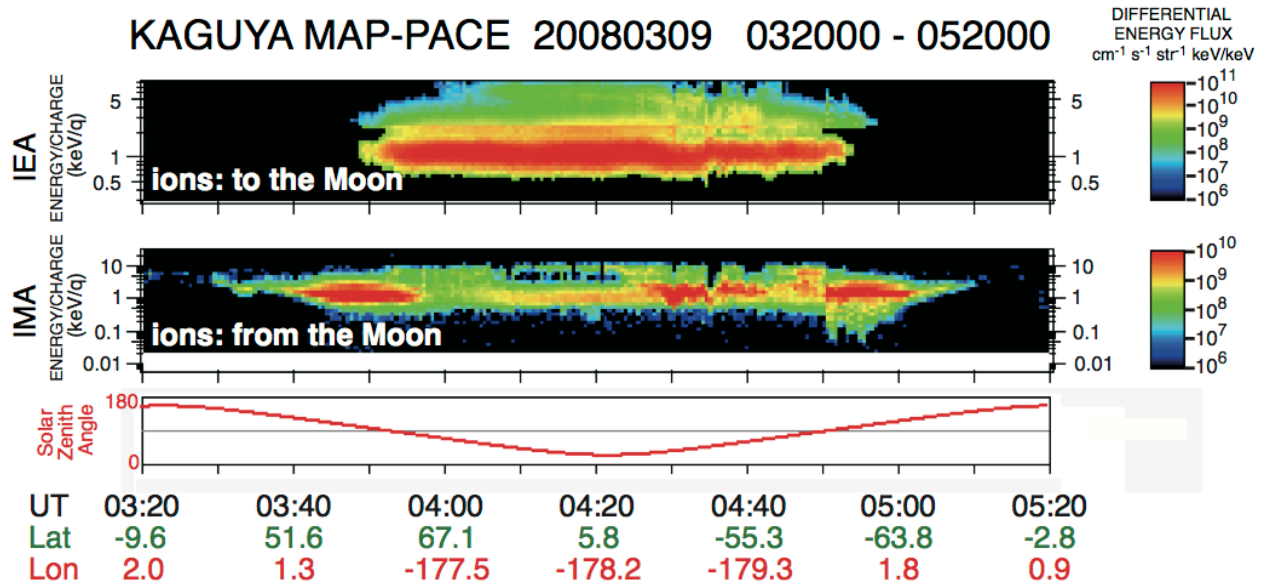
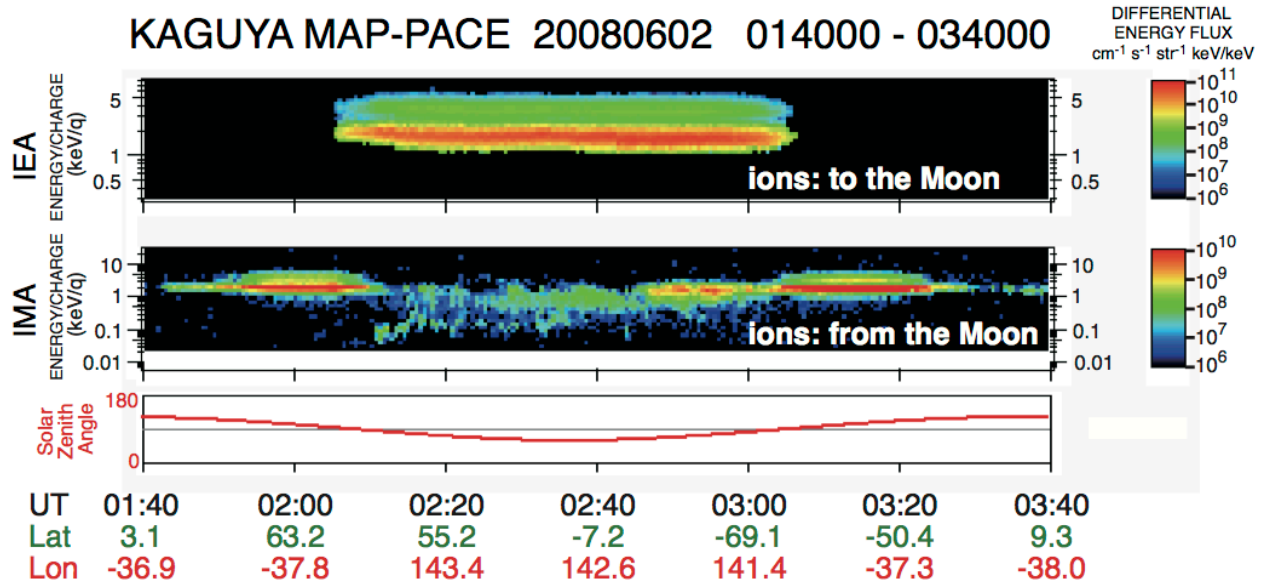
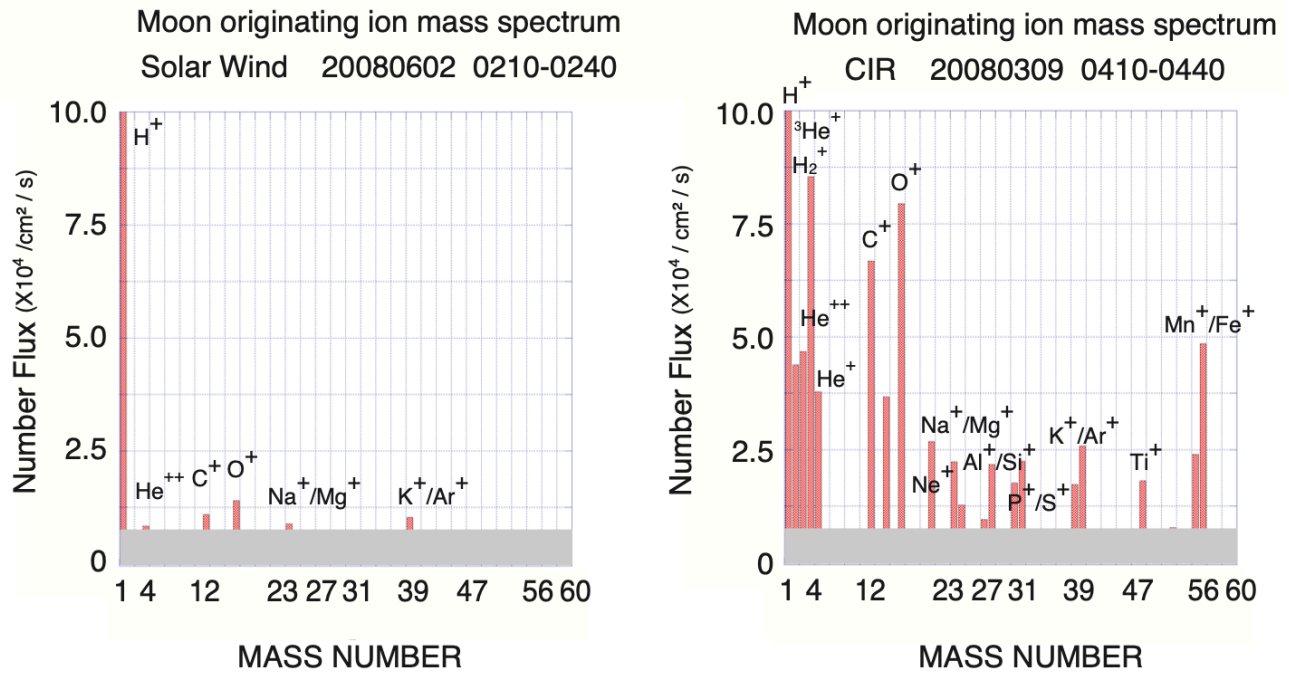


Figure 2.8: Energy-time spectrogram of ions observed by MAP-PACE on Kaguya during (a) normal solar wind condition on 2 June 2008, and (b) CIR on 9 March 2008. Moon is located upstream of the bow shock in the subsolar region. Ions measured by IEA (without mass separation) looking above the spacecraft (injection to the Moon surface) and Ions measured by IMA (with mass separation) looking downward the spacecraft (emission from the Moon surface) are shown.



1360

1361 Figure 2.9: Number fluxes of ions coming from the lunar surface for the same days as
 1362 Figure 2.8. Left: during a normal solar wind condition (2 June 2008, Figure 2.8a). Right:
 1363 during a CIR (9 March 2008, Figure 2.8b). Low charge-state indicates that they are
 1364 sputtered ions from the lunar surface rather than reflected solar wind. In both cases
 1365 Kaguya was located upstream of the bow shock.
 1366

1367

1368 The Moon-origin ions were also detected even when the Moon stayed in the Earth's
 1369 magnetosphere lobes (Tanaka et al., 2009), where the direct impact of the solar wind is
 1370 less pronounced. These ions were observed on the dayside of the Moon, especially when
 1371 the solar zenith angle was below 40 degrees. IMA detected peaks of flux for the heavy
 1372 ions including C^+ , O^+ , Na^+ , K^+ , and Ar^+ . These ions were mostly accelerated by the
 1373 convection electric field in the Earth's magnetotail. The ions originating from the lunar
 1374 surface and the exosphere showed characteristic variation of the flux intensity that
 1375 presumably related to the lunar surface structure or composition.
 1376

1377

1378 When the Moon stayed in the Earth's magnetosphere, during a high geomagnetic activity
 1379 period, the IMA instrument detected both lunar O^+ ions, originating from the Moon surface,
 1380 and energetic O^+ ions originating from the Earth's ionosphere and streaming downtail
 1381 (Terada et al., 2017). These two O^+ populations are clearly distinguished from their
 distribution functions and their energy spectra, the terrestrial O^+ ions streaming downtail

1382 with energies of the order of few keV whereas the lunar O⁺ ions have energies of the order
1383 of ~10 eV.

1384

1385 With the ion electrostatic analyzer on board the Acceleration, Reconnection, Turbulence
1386 and Electrodynamics of the Moon's Interaction with the Sun (ARTEMIS) mission, lunar
1387 pickup ions were observed when the Moon was within the terrestrial magnetotail lobe
1388 (Halekas et al., 2012; Poppe et al., 2012). Although ARTEMIS does not have any
1389 instrument that can separate the mass, it is possible to infer the existence of heavy ions if
1390 accurate ion and electron densities can be obtained. Assuming that the density calculation
1391 is accurate, Zhou et al. (2013) compared electron density and ion density when ARTEMIS
1392 detected similar energy-time profiles as Kaguya (lower energy from the solar wind or lobe
1393 plasma and the thermal component does not exist) during high flux periods, and found that
1394 the calculated ion density (assuming proton) is 5 time higher than the electron density
1395 most of the time, suggesting a significant fraction can possibly be consisting of heavy
1396 species.

1397

1398

1399 **2.4.4. Magnetospheric observation of possible Moon-origin ions**

1400 Thus, the very heavy ions of the lunar origin (sputtered ions/neutrals from the surface or
1401 ionized exospheric neutrals) constantly exist in the "lunar ion wake" (downstream plasma
1402 region of the Moon where pickup heavy ions may reach, and is wider than the lunar wake
1403 in the flow dynamical meaning due to the finite gyroradius of heavy ions). The generated
1404 heavy ions are accelerated by the solar wind convection electric field and are finally picked
1405 up, with their flux varying depending on the solar wind conditions. The question is then
1406 how much these ions contribute to the metallic ions in the magnetosphere, such as those
1407 detected by Geotail/STICS (cf. Figure 2.1). Here, we dismiss the case when the Moon is
1408 located in the Earth's magnetotail because we do not expect the Moon-origin ions to return
1409 to the Earth against the strong anti-sunward plasma flow at the Moon location, as is
1410 confirmed by Geotail/STICS (Christon et al., 2020).

1411

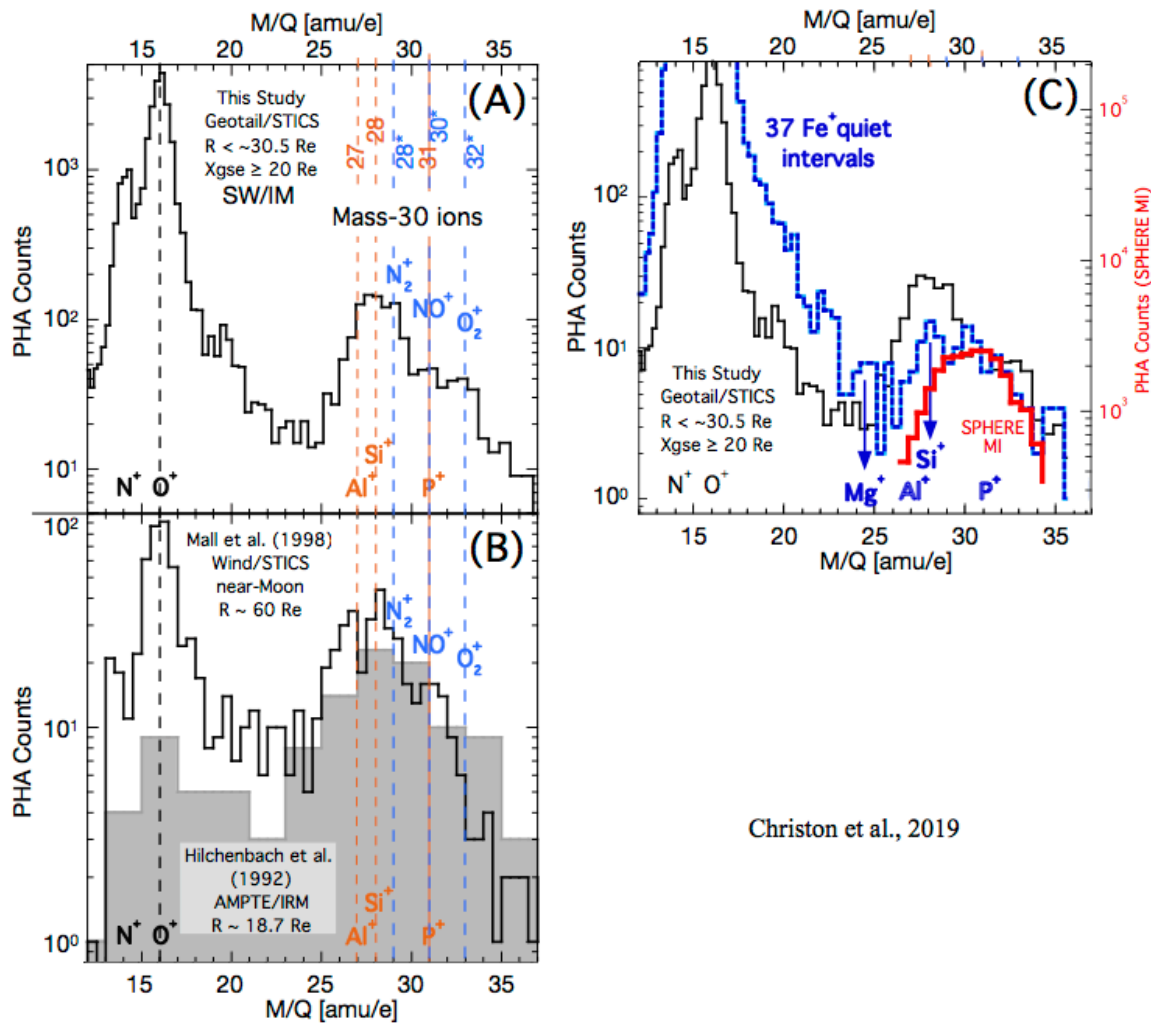
1412 Therefore, we consider the case when the Moon is located upstream of the Earth, or more
1413 precisely, when the magnetosphere is within the "lunar ion wake" (downstream plasma
1414 region of the Moon where pickup heavy ions may reach, and is wider than the lunar wake
1415 in the flow dynamical meaning due to the finite gyroradius of heavy ions). Then, the ~60

1416 Re distance from the Earth is far enough for these pickup ions to gain the solar wind
1417 speed even for Fe^+ with its large gyroradius, reaching to nearly 100 keV for Fe^+ .
1418 Considering further energization in the bow shock and magnetosheath, the other metallic
1419 ions with less mass may also reach the energy detectable by the Geotail/STICS and
1420 Wind/STICS instruments.

1421
1422 Metallic ions that are consistent with the Moon-origin ions (also consistent with Earth's
1423 origin from the metal layer) are actually detected upstream of the Earth. With the STICS
1424 instrument on the WIND spacecraft, low charge-state heavy metallic ions were observed
1425 during flybys on the earthwards side of the Moon as close as 17 lunar radii well in front of
1426 the Earth's bow shock (Mall et al., 1998). The ion composition measurements in the
1427 energy range of 20 – 200 keV/q show O^+ , Al^+ , and Si^+ ions and heavier ions. The Active
1428 Magnetospheric Particle Tracer Explorers (AMPTE) SULEICA instrument detected ions in
1429 the m/q range of 23-37 were observed in the solar wind upstream of the Earth's bow shock
1430 (Hilchenbach et al., 1993), with ion fluxes of at least $0.3 \text{ cm}^{-2} \text{ sec}^{-1} \text{ sr}^{-1} \text{ keV}^{-1}$ in the
1431 energy range of 5 keV/q to 230 keV/q.

1432
1433 Figure 2.10 summarizes these upstream observations (by Wind/STICS and
1434 AMPTE/SULEICA) and compared with the Geotail/STICS observation. Although the
1435 Geotail/STICS count rate of these low charge-state metallic ions is, including all sources,
1436 normally 0-1 count/day, the operating lifespan of more than 20 years and its $\sim 9 \times \sim 30 \text{ Re}$
1437 orbit allowed statistical studies of near-Earth heavy ions in the magnetosphere, the
1438 magnetosheath, and the solar wind sunward of Earth. In Figure 2.10 (cf. Christon et al.,
1439 2020), the peak mass around $m/q \approx 30$ is substantially different between the upstream
1440 data (black trace in (C), around mass 27 - 28: Al^+ and Si^+) and magnetospheric data (red
1441 trace in (C), around mass 30 - 32: NO^+ and O_2^+). Unlike the magnetospheric peak that is
1442 dominated by the molecular ions (N_2^+ , NO^+ , O_2^+), the upstream peak is centered at Al^+ and
1443 Si^+ (slightly lower mass than molecular ions) in good agreement with mass spectra
1444 observed by Kaguya and LADEE (Saito et al., 2010; Halekas et al., 2015) as described in
1445 Sect. 2.4.

1446



Christon et al., 2019

1447

1448 Figure 2.10: <<energeticMass-30_Lunar_Ions.pdf>>

1449 Histograms of ion pulse height analysis (PHAs) events ordered by m/q with the ion species
 1450 N⁺, O⁺, and ions around mass 30 (Christon et al., 2019, Fig 9). (A) Geotail/STICS data
 1451 (~87 - 212 keV/q) at farthest upstream (XGSE = 20 - 30.5 Re). (B) Solid line: Wind/STICS
 1452 data (~20 - 200 keV/q) near the Moon at >17 lunar radii when Wind is at sunward of the
 1453 Earth (Mall et al., 1998), and shaded area: AMPTE/IRM data (80 - 226 keV/q) upstream of
 1454 the bow shock at 18.7 Re from (Hilchenbach et al., 1993). Vertical orange dashed lines,
 1455 which pass through the peaks of solid lines (both Geotail and Wind data), correspond to
 1456 the masses for light metallic ions of lunar origin: 27 (Al⁺) and/or 28 (Si⁺), and 31 (P⁺) and/or
 1457 32 (S⁺). As reference, mass lines for molecular ions (N₂⁺=28, NO⁺=30, O₂⁺=32) are shown
 1458 by vertical blue dashed lines and mass line for O⁺ is shown with black dashed line. Geotail
 1459 data were obtained over approximately 2 full solar cycles, Wind data were obtained during
 1460 tail traversals over 1995 - 1997 (solar minimum), and AMPTE data were obtained over 3
 1461 months in late-1985 (solar minimum). (C) Geotail/STICS data at, black line: farthest
 1462 upstream (the same as (A)); red line: the overall magnetosphere data (SPHERE in Figure
 1463 1.1) which is dominated by molecular ions; and blue line: the average of 37 orbits (data
 1464 over 24 hr covering all regions) during low to moderate solar/geomagnetic condition. The
 1465 data are translated vertically to match values at ≥ 30 - 32 amu/e.

1466

1467

1468

1469 However, the existence of low charge-state heavy ions does not necessarily mean that the
1470 excess part of upstream counts beyond the magnetospheric profile ($m/q = 22-30$) is of
1471 lunar origin, because (1) in addition to N_2^+ and CO^+ ($m/q = 28$), metallic ions at this mass
1472 ranges exist in the Earth's upper atmosphere (Na^+ (23), Mg^+ (24), Al^+ (27), and Si^+ (28))
1473 through the ablation of meteoroids (Plane et al., 2016) that forms the
1474 mesospheric/thermospheric metal layer, as described in Sect. 2.3, (2) their large gyroradii
1475 makes it possible to access the far upstream region by the foreshock (Kronberg et al.,
1476 2011) once they reach the space, although lifting these very species to the exobase is
1477 extremely difficult (Schunk and Nagy, 2009), and (3) the solar wind may also contain low
1478 charge-state heavy ions through the solar wind comet/microdust interaction. Thus, the
1479 mass profiles cannot be used to select the lunar ions detected even in the solar wind
1480 upstream of Earth (Christon et al., 2019). We come back this problem in §4.1.

1481

1482

1483

1484 **3. List of datasets and models that actually detected molecular/metallic** 1485 **ions**

1486

1487 As summarized in Section 2, observations of molecular and metallic ions and modelling of
1488 their transport are important from many inter-disciplinary aspects. However, these very
1489 minor ions are vastly unexplored in the near-Earth space. This is because only a few
1490 terrestrial missions have been equipped with dedicated instrumentation capable of
1491 separating these molecular and metallic ions, and because even these few dedicated
1492 instruments were capable of detecting a limited energy range (cold ions of < 50 eV and
1493 energetic ions of ~ 100 keV) and a limited mass range (≤ 40 amu). Nevertheless, existing
1494 data from the past and on-going missions, including those not designed for the required
1495 mass separation, are sometimes capable of detecting some of these molecular (very
1496 limited for metallic) ions with available tools, although severe limitations exist (sensitivity
1497 and energy range, in addition to mass resolution and mass range). In this section, we list
1498 these datasets.

1499

1500

1501 **3.1 Satellite datasets**

1502 Table 3.1 summarizes the available datasets from the magnetospheric and Moon missions
1503 by which the heavy molecular or metallic ions are observed. As a reference, we also list
1504 the capability of planetary missions with the Earth flyby data in Table 3.2. The table
1505 includes the measurement methods because they determine the approximate resolution,
1506 sensitivity, and mass-energy ranges. There are roughly three different methods of mass
1507 separation for particle instruments: magnetic method, time-of-flight (TOF) method in
1508 various formats, and retarding potential analyzer (RPA) method. By combining with the
1509 electrostatic analyzer for energy per charge (E/q) selection at the entry of the instrument
1510 (by using a perpendicular electric field, only ions with specific energy per charge can follow
1511 the curved entry), the magnetic method and the TOF method give m/q and E/q ,
1512 respectively.

1513

1514

1515

1516

1517

Table 3.1: Terrestrial missions that reported heavy molecular or metallic ions since 1980's

mission/instrument (duration) @where	method	Specification (*1)	what was actually observed	reference (spec + obs m>26)
Cluster/CIS (2001-) @magnetosphere (almost everywhere)	TOF (for > 28 eV) RPA (for low-energy)	0.7 eV/q – 40 keV/q m/q: 1 - 16 (H ⁺ , He ⁺⁺ , He ⁺ O ⁺)	1 - ~40 amu/q (~50 lower statistics) m/Δm ~ 5 - 7	Rème et al., 2001. Kistler et al., 2013.
Cluster/RAPID (2001-) @magnetosphere (almost everywhere)	SSD TOF	> 400 keV H, He, and O (C, N, O are not separated)	m>16 up to more than 60	Wilken et al., 2001. Haaland et al., 2020, Haaland et al., 2021.
e-POP/IRMS (2013-2021) @325-1500 km altitude	TOF	< 70 eV/q m/q: 1-64	N ⁺ and O ⁺ are separated but not between N ₂ ⁺ , NO ⁺ , and O ₂ ⁺ .	Yau et al., 2015. Yau et al., 2016.
AraSe/LEPI (2017-present) @inner magnetosphere	TOF	0.01-25 keV/q m/q: 1 - 40	H ⁺ , He ⁺⁺ , He ⁺ , O ⁺⁺ , O ⁺ , O ₂ ⁺ (no separation between N and O) m/Δm ~ 2-4	Asamura et al., 2018. Seki et al., 2019. (statistics)
AraSe/MEPI (2017-present) @inner magnetosphere	SSD TOF	7 - 87 keV/q m/q: 1 - 48	H ⁺ , He ⁺⁺ , He ⁺ , O ⁺⁺ , O ⁺ , O ₂ ⁺ (no separation between N and O) observed m/Δm ~ 3-5	Yokota et al., 2017. Seki et al., 2019. (statistics)
Kaguya/MAP-PACE/IMA (2008-2010) @Moon	LEF-reflectron	10 eV/q – 28 keV/q m/q: 1 - ~ 60 m/Δm ~ 5 (m/Δm ~ 15 for reflected ions < 12 keVq)	Na ⁺ , Ca ⁺ , K ⁺ , C ⁺ , Si ⁺ , O ⁺ are separated. no molecular ions (due to start foil breaks molecules)	Saito et al., 2010 Yokota et al., 2009.
Geotail/STICS (1992-2022) @magnetosphere	SSD TOF	energy > 200 keV/q for m=30 m: <1 up to ~60-70 amu m/q: <1 up to ~95 amu	10 ⁶ -2 Fe ions/3hr in -> at best once every 900 hr	Christon et al., 2017.
Wind/STICS (1994-) @magnetotail	SSD TOF	6 - 230 keV/q otherwise, the same method as Geotail.	Separate O ⁺ from Al ⁺ , Si ⁺	Mall et al., 1998.
Polar/TIDE (1996-2008) @below 8 Re	TOF	0.1-500 eV/q 1 - 40 amu/q m/Δm = 4	"Significant component of molecular ions in polar wind flux in response to CME"	Moore et al., 1995.
Polar/TIMAS (1996-2008) @below 8 Re	Magnet (double focusing)	15 eV/q – 33 keV/q m: 1 - >32 amu/q m/Δm = 2-5	Separate O ₂ ⁺ from O ⁺ (but not between N ₂ ⁺ , NO ⁺ , and O ₂ ⁺)	Shelley et al., 1995. Lennartson et al., 2000.
Akebono/SMS (1989-2002) @up to 10000 km	RPA	< 50 eV/q m/q: 1 - 40	mostly <20 eV Sometimes separate among N ₂ ⁺ , NO ⁺ , and O ₂ ⁺	Whalen et al. 1990. Yau et al., 1993. Yau et al., 1998.
AMPTE/CHEM (1984-1989) @inner magnetosphere	TOF	1-300 keV/q m: <1 up to 90 m/q: <1, 74	Can separate N ₂ ⁺ , NO ⁺ , and O ₂ ⁺	Gloeckler and Hamilton, 1987.
AMPTE/SULEICA (1984-1986) @inner magnetosphere	TOF	< 50 eV/q	Detected metallic ions (SW charge state)	Stern, 1999. Hilchenback, 2004.
DE-1/RIMS (1981-1984) @568km x 4.6 RE	RPA	10 eV/q - 17 keV/q m/q = 1 - 150 m/Δm ~10	Can separate N ₂ ⁺ , NO ⁺ , and O ₂ ⁺	Chappell et al., 1981. Craven et al., 1985.
DE-1/EICS (1981-1991) @568km x 4.6 RE	Magnet		O ₂ ⁺ , NO ⁺ , N ₂ ⁺ separated (in 'drum mode')	Shelley et al. 1981.
MMS/HPCA (2015-present) @Magnetosphere	TOF	1 eV/q - 40 keV/q m/q: 1 - 16 (H ⁺ , He ⁺⁺ , He ⁺ , O ⁺⁺ , O ⁺)	Fe ⁶⁺⁻⁷⁺ by chance but not by search	Young et al., 2016. Gomez et al.,

(*1) energy is with respect to spacecraft potential

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(*1) energy is with respect to spacecraft potential

1522 Table 3.2: Planetary missions capable of separating metallic ions and molecular ions of <
 1523 100 keV

mission/instrument with Earth flyby data	metallic ions	molecular ions from atomic ions	Earth flyby?	reference
STEREO/PLASTIC	TOF	0.2-80 keV up to Fe	molecular @Tail 200-300 R _E (2007)	Galvin et al., 2008 Kistler et al., 2010b
Cassini/MIMI-CHEMS	TOF	~10 – 220 keV m: 1 - 80	only ring current ions @ring current (1999)	Christon et al., 2017
JUICE/JDC (Launched 2023)	TOF	1 eV – 35 keV m: 1 - 70 m/Δm=30	Planned: 1 Moon (< 300 km) + 3 Earth (< 10000 km) flybys	Withmann, 2022
JUICE/NIM (Launched 2023)	LEF-reflection	< 10 eV (both neutrals + ions) m: 1 - 1000 m/Δm=800	same as above	Föhn et al., 2021

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1525

1526 The TOF method has many variants: foil-type for energetic particles (e.g. Wilken et al.,
1527 2001), combination of an electrostatic analyzer with a foil-type TOF unit (e.g. Rème et al.,
1528 2001), combination of an electrostatic analyzer with a linear electric field embedded TOF,
1529 so called LEF-reflectron unit (e.g. Delcourt et al., 2016), combination of an
1530 ELECTROSTATIC ANALYSER with a grazing incidence microchannelplate (MCP) TOF
1531 (e.g. Devoto et al., 2008), combination of an ELECTROSTATIC ANALYSER with a gated
1532 TOF unit (e.g. Keller et al., 1999), combination of an ELECTROSTATIC ANALYSER with a
1533 reflecting surface TOF unit (Wittman, 2022), and straight start-end pair for a simple TOF.
1534 For hot plasma of 0.05 - 10 keV energy range, the LEF-reflectron so far provides the
1535 highest mass resolution $m/\Delta m$. The details of the different measurement methods can be
1536 found in a review paper (Wüest et al., 2007). As a general problem with the TOF method
1537 using start foil or start surface, some molecular ions are dissociated into atomic ions and
1538 atoms there, making the TOF spill out toward longer TOF from the molecular ion peak.
1539 This is why the Kaguya data does not include molecular ions. Nevertheless, TOF is the
1540 most liable method to separate the mass.

1541

1542 Separation of species within the same mass group (between C, N, and O; between N₂,
1543 CO, NO, and O₂) is another important challenge (particularly separation of N and O), but
1544 that requires $m/\Delta m > 50$ (we call instruments with such capability as mass spectrometer).
1545 The same or even better is required for the detection of metallic ions because of the
1546 much lower flux compared to the molecular ions.

1547

1548 As summarized in Table 1.1, there is a clear lack of mass spectrometers covering
1549 molecular and metallic ions for < 10 keV for near-Earth missions. However, mass
1550 spectrometers are regularly included in many deep-space missions, the Moon missions,
1551 and solar wind monitoring spacecrafts (like ACE: advanced composition explorer). Among
1552 them, Earth-flyby data exist for some mission such as Stereo. The Earth-flyby itself
1553 occurred for more missions such as Bepi-Columbo (MPO and MMO), Solar Orbiter,
1554 MAVEN, and Rosetta, but heavy ion data were not taken or not useful.

1555

1556 Although excluded from Table 3.1, heavy molecular ions (O₂⁺) might be able to be
1557 separated from the atomic ions (O⁺) by a simple electrostatic analyzer when ions are
1558 streaming with a group velocity, such as the far downstream of the Earth. In such cases,
1559 different masses correspond to different kinetic energies, and hence the different ion

1560 species appear as different groups in an energy-time spectrogram. SOHO/CELIAS (in the
1561 Venus magnetotail at 0.3 au) detected heavy ions of Venus tail origin, and well separated
1562 C^+ and O^+ (Grünwaldt et al., 1997) by carbon-foil time-of-flight (C-TOF). This kind of rough
1563 mass separation has been tried for the ion instrument onboard the THEMIS-ARTEMIS
1564 spacecraft, which are in a lunar orbit, when the Moon is within the terrestrial magnetotail
1565 and detects downstream ions of terrestrial origin. It allowed the separation of H^+ from
1566 heavy ions (Poppe et al., 2016a). However, the possible O_2^+ signature was not separated
1567 from the O^+ signature. To separate them, all ion species must be downstreaming with
1568 exactly the same velocity, but this hypothesis breaks down at large distances since heavy
1569 ions tend to reach higher flow velocities than protons (Seki et al., 1998).

1570

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1572 **3.2 Analyses tools to extract molecular/metallic ions in the space missions**

1573

1574 Table 3.3 summarizes the analyses tools of these data to extract the molecular and
1575 metallic ions. Note that some analyses tools might not be working with modern computer
1576 environments.

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Table 3.3: Access method of relevant dataset and software with note on availability missions (everybody fill data type and software)

mission /instrument	@where	database	notes on the software (basically request-basis)
Cluster/CIS	Magnetosphere	ESA Cluster Science Archive (ASCII) for routine product: https://csa.esac.esa.int/csa-web/ A separate telemetry product ("selected TOF events") for TOF. ESA Cluster Science Archive (ASCII) for near-raw product: https://csa.esac.esa.int/csa-web/ ePOP-data.phys.ucalgary.ca	IFSI TOF software (IDL source code): Reads level-1 binary files (selected TOF events).
Cluster/Rapid	Magnetosphere		Direct event (TOF)
e-POP/IRM	325-1500 km altitude		Reduced TOF (software for general-user access under development)
Arase/LEPI Arase/MEPI	@ring current and inward	ERG science center (CDF file): https://ergsc.isee.nagoya-u.ac.jp/data_info/erg.shtml.en	(1) IDL SPEDAS for main data products, omcluding TOF histograms. (2) IDL code for MEPI to look at raw PHA data
Kaguya/MAP- PACE IMA	Moon	SELENE data archive	(1) C code by Y. Saito (read binary data) (2) IDL code Y. Harada
Wind/STICS	Magnetotail	NASA cdaweb (web interface) and spdf (CDF file) https://spdf.gsfc.nasa.gov/pub/data/wind/sms/I2/	
Polar/TIDE	Magnetosphere below 8 R _E	Level-Zero Telemetry Files hosted by the GSFC https://spdf.gsfc.nasa.gov/pub/data/polar/fips://pwgdata.gsfc.nasa.gov/pub/compressed/pol/tid/	
Polar/TIMAS	Magnetosphere below 8 R _E	POLAR TIMAS H1 and H2 high resolution data: https://lasp.colorado.edu/timas/info/h12-data/h12-data.html Summary data: https://lasp.colorado.edu/timas/data/summary/	https://lasp.colorado.edu/timas/info/h12-data/make_h2.pro https://lasp.colorado.edu/timas/info/h12-data/make_h2_cdf.pro
Geotail/STICS	Outer magnetosphere	summary plots available at http://sd-www.jhuapl.edu/Geotail/Years_dir.html	code developed by S. Nyland and S. Christon
Akebono/SMS	Magnetosphere below 10000 km	JAXA/ISAS Data Archives and Transmission System (DARTS): https://darts.isas.jaxa.jp/stp/akebono/SMS.html	under conversion to public database
AMPTE/CHEM	Inner magnetosphere	APL AMPTE site: http://sd-www.jhuapl.edu/AMPTE/chem/index.html NASA spdf site: https://spdf.gsfc.nasa.gov/pub/data/amppte/ccel/	(1) Python code at UNH reading the CHEM FITS files (2) PHAFLUX fortran code calculates fluxes from PHA events
AMPTE/SULEICA	Inner magnetosphere	data is not easy to access	
DE-1/RIMS	Inner magnetosphere	NASA spdf site: https://spdf.gsfc.nasa.gov/pub/data/de/1/plasma_rims/	summary spectra https://spdf.gsfc.nasa.gov/pub/data/de/de1/plasma_rims/de1_rims_summary-spectrograms_nasa-tm-19950009193.pdf
DE-1/EICS	Inner magnetosphere	NASA spdf site: https://spdf.gsfc.nasa.gov/pub/data/de/1/particles_eics/	
MMS/HPCA	equatorial magnetosphere	NASA cdaweb and spdf site: https://spdf.gsfc.nasa.gov/pub/data/mms/mms1/hpca/srvy/I2/tof-counts/ https://spdf.gsfc.nasa.gov/pub/data/mms/mms2/hpca/srvy/I2/tof-counts/ so on	TOF at NASA cdaweb https://cdaweb.gsfc.nasa.gov/cgi-bin/eval1.cgi

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1583 3.3 Mesospheric and ionospheric dataset including sounding rocket

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1585 As the source region's information, datasets for the metal layers and ionospheric
 1586 molecular ions are useful. Since they can be observed from the ground and sounding
 1587 rocket in addition to some spaceborne observations (e.g., limb scanning), there are many
 1588 databases, as summarized in Sect 2.3.2. Table 3.4 summarizes observation methods with
 1589 some examples. We just show examples for the databases because most of them are not
 1590 publicly available or the database is not internationally organized

1591

1592 Table 3.4: Available method of monitoring the uplift of the molecular and metallic ions in the
 1593 ionosphere and mesosphere

method	where	examples	species	data availability
Resonance-Scattering Lidar	mesosphere - 80 km up to 200 km (only sometimes Na)	ALOMAR (1*): Tromsø for Na (*2):	Fe, Na, Ni, K, Ca (Mg cannot be observed at 285 nm because of the stratospheric ozone layer) Na from 2010- 2019 at Tromsø	Plane et al. (2015) CEDAW database (http://cedar.openmadrigal.org/) has list but no "open" data site. Data (Na) at Tromsø upon request
ion/neutral mass spectrometer (sounding rocket)	mainly E-layer below 120 km	total < 10 for metallic ions (*3)	few paper/rockets, but not more knowledge than Lidar data (Plane's paper)	Kopp (1997) Grebowsky and Aikin (2002)
ion/neutral mass spectrometer (satellite)	< 30° lat < 400 km alt	Ogo 6, AE-C, AE-D,	molecular results are already included in standard model	
Satellite: limb optical	< 400 km alt	Odin/OSIRIS, (TIMED), Envisat/SCIAMACHY	Mg ⁺ , Mg, Na and K	On request to University of Leeds (W. Feng)

1594 *1: <https://www.iap-kborn.de/en/research/department-optical-soundings-and-sounding-rockets/instruments-and-models/metal-lidar-kuehlungsborn/>

1595 *2: <https://www.isee.nagoya-u.ac.jp/~nozawa/indexlidardata.html>

1596 *3: One example from molecular detection, one example from metallic detection, and example of
 1597 obtaining extra information from Lidar observation of > 130 km

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1601 3.4 Modelling of contribution from meteor and space debris through deposition to 1602 the metal layer

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1605 Table 3.5: Models relevant to upflow of molecular and metallic ions reaching the exobase

Name	Description	Citation/availability
SAMI-3	Predict metallic ion in the ionosphere (Huba's model includes transport to 600 km)	
WACCM	Metallic ions in the ionosphere.	Feng et al., 2013; Wu et al., 2021
CABMOD	Examine differential ablation of meteoroids and deposition as a function of altitude and orbital position	Bones et. al. 2019, Vondrak et. al. 2008, Carrillo-Sanchez et. al. 2020. Data available on request ^(*) .
WACCM-X	Numerical model spanning the range of altitude from the Earth's surface to the upper thermosphere.	Wu et al., 2021 www2.hao.ucar.edu/modeling/waccm-x
ORSAT	Predict the re-entry survivability of satellite and launch vehicle upper stage components.	Dobarco-Otero et. al. 2005 orbitaldebris.jsc.nasa.gov/reentry/orsat.html
SCARAB	Simulate the re-entry of a satellite in detail.	Koppenwallner et. al. 2005 www.htg-gmbh.com/en/htg-gmbh/software/scarab/
MASTER	Assess the debris or meteoroid flux imparted on a spacecraft on an arbitrary earth orbit.	Flegel et. al. 2009 sdup.esoc.esa.int/
LEGEND	Three-dimensional debris evolutionary model for long-term debris environment projection.	Liou et.al 2004 orbitaldebris.jsc.nasa.gov/modeling/legend.html
-	A dynamical model of the sporadic meteoroid complex.	Wiegert et al., 2009
-	Fluid model (simulation) in the ionosphere	Shinagawa and Oyama, 2006
	Polar wind outflow model for < 300 km – > 8000 km	Glocer A (GFSC/Mishigan)

*1 Request to Juan Diego Carrillo Sanchez (juandiego.carrillosanchez@nasa.gov)

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Table 3.5 summarizes models relevant to upflow of molecular and metallic ions reaching the exobase. The table also includes models of space debris as these could contribute to the mesospheric metal layer if their ablation altitudes are high enough. These models provide the distribution of the modeled species beyond what the empirical International Reference Ionosphere (IRI) model provides.

For the ionospheric and mesospheric heavy ions to be lifted, a common model of the neutral convection and electromagnetic fields to estimate the dynamics of the atomic ions can be used for both the molecular and metallic ions. Note that the available models still underestimate the upward convection in the polar region (see Shinagawa and Oyama,

1619 2006). Even the solar flare effect, i.e., the heating of the ionosphere by the flare-related
1620 radiation, cannot reproduce the extremely high convection that is observed (Yamauchi et
1621 al., 2020). Still the existing models are a good start to estimate the upper limit and lower
1622 limit of the flux of metallic ions into the magnetosphere.

1623

1624 Since the ionospheric metal layer is most likely formed by the ablation of meteoroids, a
1625 current update to the ablation modeling is relevant. Then, we need an empirical model of
1626 lifting (expanding) the metal layer to the ionospheric altitude (up to > 120 km) such that an
1627 ionospheric/thermospheric model of upflow can be applied. Finally, we note that it is
1628 difficult to model once the molecular and metallic ions reach the exobase, because wave
1629 activity is required to lift them to the region where sufficient energization is expected.

1630

1631

1632 **4. Merit of combining data from different sources and models**

1633

1634 In this section, we show some examples of the merit of combining data and knowledge
1635 from different sources.

1636

1637 **4.1 Moon contribution to energetic (> 100 keV) ions**

1638

1639 As summarized in Sect. 2.4, the mass spectrum of the very heavy ions in the Earth's
1640 upstream region while downstream of the Moon is consistent with that coming from the
1641 Moon. However, this is yet not sufficient to state that majority are of lunar origin, as
1642 discussed in Sect. 2.4.4. We revisit this problem by combining the Geotail/STICS
1643 observation with the other data such as the Moon location and the solar wind monitor.

1644

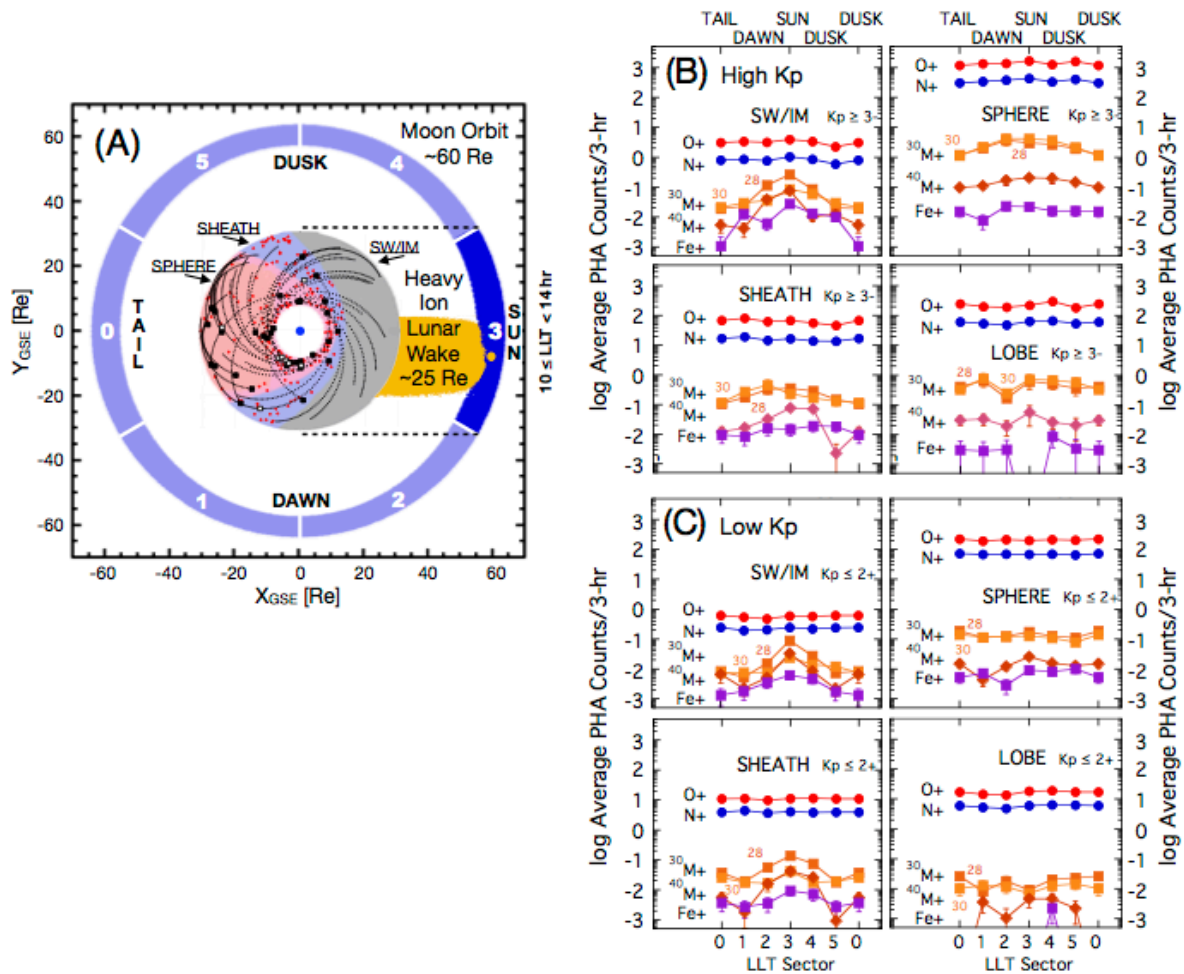
1645 **4.1.1 Statistics of Energetic ions (Geotail)**

1646 Christon et al. (2020) examined the Geotail/STICS counts in term of the Moon location. To
1647 overcome the low count-rate problem (0-1 count per day), they integrated the data over
1648 nearly 20 years of Geotail data, and further integrated the data over the location for both
1649 the Geotail (four regions which are the same as Figure 2.1) and the Moon (six locations:
1650 the upstream near the new Moon, four sides, and the downstream near full Moon). Figure
1651 4.1 shows the results. For example, sector 3 in Figure 4.1a shows the result of collecting
1652 all cases when the Moon is located sunward of Geotail's nominal orbital XGSE-YGSE
1653 range.

1654

1655 Figures 4.1b and 4.1c (Christon et al., 2020, Fig. 7) show the results for high K_p (≥ 3) and
1656 low K_p (≤ 2) cases, respectively. For both K_p cases, heavy ion counts (for $m/q \geq 28$, i.e.,
1657 molecular, metallic ions) in the upstream solar wind (SW/IM in the figure) show a peak at
1658 LLT sector 3 (5-day period near the new Moon), with a widening of the peak for higher K_p .
1659 This peak at LLT sector 3 is less obvious for the magnetosheath (SHEATH in the Figure
1660 4.1) and there is nearly no peak at sector 3 in the magnetosphere (SPHERE) and plasma
1661 lobe (LOBE). Considering that heavy ion flux of $m/q \geq 28$ escaping from the Earth (almost
1662 all are molecular ion) is nearly zero for low K_p , all these results indicate that Moon origin
1663 metallic ions are the major heavy ($m/q > 20$) ions in the upstream region. Here, the
1664 increase of the ion counts for high K_p (which generally means stronger solar wind) agrees
1665 with the general increase of both the Earth-origin molecular ion flux (not dependent on the

1666 Moon location, and seen in all panels) and the Moon origin heavy ion flux (peak at LLT
 1667 sector 3).
 1668



1669
 1670 Figure 4.1: <<Lunar ion wake and local time.pdf>>

1671 (A) A sketch of the Earth (blue dot at the center), the Moon's orbital range (~60 Re), and Geotail
 1672 orbital range (~9 < R < 35 Re). Two spatial criteria for considering possible lunar pickup ion
 1673 influence in Geotail/STICS suprathermal (~87 - 212 keV/e) ion measurements are the lunar local
 1674 time (LLT) and the "Lunar-ion Wake". LLT marks the orbital location of the Moon with respect to
 1675 the Earth-Sun line. For example, 10 hours ≤ LLT ≤ 14 hours (the sector marked as 3 in the figure)
 1676 corresponds to the Moon location sunward of Geotail's nominal orbital X_{GSE} -Y_{GSE} range. The
 1677 "Lunar-ion Wake" (different from fluid dynamical "wake") drawn here (~25 Re width) is the region
 1678 where very heavy ions of lunar origin (e.g., CO²⁺ or Fe⁺) are expected exist in the nominal IMF at
 1679 1 au (~7 - 9 nT). Selected segments of Geotail orbits (dotted traces near Earth) terminate when an
 1680 Fe⁺ was observed during low to moderate solar and geomagnetic conditions. White (black)
 1681 squares indicate Fe⁺ observations obtained when the Moon was (not) in LLT-sector 3. Red dots
 1682 show other measured Fe⁺ data. Three different regions (magnetosphere: "SPHERE",
 1683 magnetosheath: "SHEATH", and upstream of bow shock "SW/IM") are identified by different
 1684 colours. The plasma lobe ("LOBE in Figure 1.1) overlies the SPHERE and not shown here. (B)
 1685 Average counts/3-hours of low charge state ions at four different regions during Kp ≥ 3 and (C)
 1686 those during Kp ≤ 2.
 1687

1688

1689 The results also indicate that unless the Moon is directly upstream of the Earth (i.e., except
 1690 LLT sector 3), the magnetospheric heavy ($m/q > 20$: metallic) ions are mainly provided
 1691 from the Earth with minor contribution from the Moon most of the time. Both contributions
 1692 can be comparable if we limit to a 5-day average near new Moons because the peak
 1693 values for $m/q > 20$ ions near LLT sector 3 are the same level between in the upstream
 1694 (SW/IM and SHEATH) and in the magnetosphere (SPHERE). The enhancement near LLT
 1695 sector 3 during high K_p suggests that there might exist periods (e.g., for $K_p \geq 5$) when the
 1696 metallic ions of lunar origin are found more than those from the Earth origin.

1697

1698

1699 **4.1.2 Case studies** (combining Geotail and ACE)

1700 To obtain a more concrete view of the Moon-origin ions in the magnetosphere with the
 1701 existing dataset of very low count rates (0-1 count per day for singly-charged metallic ions
 1702 of mass more than 33), we need to carefully select the conditions when the solar wind flux
 1703 significantly increased while the Moon is upstream. Here we show the usefulness of
 1704 combining different datasets (Geotail, ACE, and SOHO) for such a study. We selected
 1705 CME-driven interplanetary shock events (from list made by SOHO/CELIAS proton monitor)
 1706 with significant flux increase of iron ion (Fe^{n+}) by three order of magnitude (to over $100 \text{ cm}^{-2} \text{ s}^{-1} \text{ str}^{-1} \text{ MeV}^{-1}$) in ACE/ULEIS instrument) within two days from new Moon. Out of them,
 1707 half-day resolution QL of Geotail/STICS is available for 10 events: 6 events when Geotail
 1708 is located within the lunar wake, and 4 events outside the lunar wake.

1710

1711 For these “best” events, we counted all half-day counts when the interplanetary shock
 1712 arrived and compared them from those when the Geotail was in the same region one orbit
 1713 before (about 5 days before). Here, the region is judged from the orbit, low-energy particle
 1714 (LEP) data and STICS data. Table 4.1 summarizes the results. The third column lists the
 1715 change in the half-day triple-coincidence counts for $m > 34$ and $m/q \geq 34$ (inside parenthesis
 1716 are additional counts that satisfies $m > 20$ and $m/q \geq 34$). For all cases, counts during one
 1717 orbit before the CME arrival (5 days before, i.e., Geotail is outside the lunar wake) are
 1718 zero. However, after the arrival of the interplanetary shock, the Geotail sometime detects
 1719 counts corresponding to the metallic ions apparently heavier than O_2^+ if and only if the
 1720 Geotail is located within the lunar wake.

1721

1722 Table 4.3: Change in the half-day integrated counts of low charge-state metallic ions
 1723 observed by Geotail/STICS.

shock timing	location ^{*3}	triple: m>34	double: m/q 39-46
2000-04-06, 16:01	SH	0 to 1 (0 to 2)	0 to 41
2000-11-26, 07:15	SW	0 to 0 (0 to 0)	0-1 to 3
2002-09-07, 15:54	SW	0 to 1 (0 to 0)	0 to 7
2003-05-29, 11:52	SH	0 to 4 (0 to 0)	2 to 42
2004-01-22, 01:10	SH to SPH	0 to 11 (1 to 8)	201 to 463
2005-09-02 ^{*1} , 13:32	SW	0 to 0 (0 to 0)	0 to 0
2000-10-28, 09:01	SW ^{*4}	0 to 2 (0 to 0)	2-4 to 2-3
2001-01-23, 10:15	SPH ^{*4}	0 to 0 (0 to 0)	243 to 193
2003-10-24 ^{*2} , 14:47	SW ^{*4}	0 to 0 (0 to 0)	0 to 0
2004-09-13, 19:29	SW ^{*4}	0 to 0 (0 to 0)	0-1 to 1-3

1724 *1: Peak of Aurigids meteor shower was 2005-08-31

1725 *2: Peak of Orionids meteor shower was 2003-10-21

1726 *3: SH: magnetosheath, SW: solar wind, SPH magnetosphere

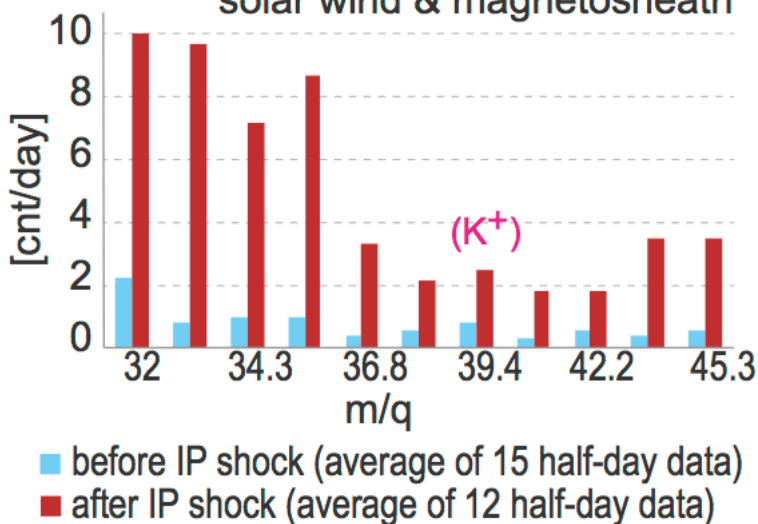
1727 *4: Geotail was not in the wake region for this time interval.

1728

1729

Geotail/STICS, double coincidence

solar wind & magnetosheath



1730

1731 Figure 4.2: <m/q_STICS.ai> Mass-per-charge (m/q) distribution of double coincidence
 1732 counts for mass ≥ 32 obtained by Geotail/STICS. Half-day data inside the solar wind or
 1733 magnetosheath before and after the interplanetary (IP) shocks with highest Fe^{n+} fluxes
 1734 listed in table 4.1 (total 8 events) are averaged. For each event, maximum two half-day
 1735 data (if staying long within the solar wind) are included. A small peak is Spread of the
 1736 major peak at around $m/q=39$ sticks out from the wide spread from $m/q \approx 32$.

1737

1738

1739 Since the majority of these cases were observed when the Geotail was in the solar wind,
1740 the result is consistent with the hypothesis that low charge-state heavy metallic ions in the
1741 upstream region seen in Figure 2.1 are mostly from the Moon when the solar wind had
1742 favorable conditions for ejecting the lunar elements (sputtering or charge exchange). This
1743 is more obvious in the double-coincidence counts for $m/q \geq 39$ during the same half-day
1744 periods (fourth column of Table 4.3). Here, we take $m/q \geq 39$ because its peak
1745 (corresponding to potassium ion K^+) is well separated from mass 32 (S^+ or P^+ rather than
1746 O_2^+ because it is most likely the lunar origin), as shown in Figure 4.2.

1747

1748 Thus, just combining the available summary data of the Geotail/STICS and a solar wind
1749 monitor can provide more insight on the metallic ions in the upstream region, and so it is
1750 worthwhile to try further combinations with the other datasets (e.g., geomagnetic activity)
1751 to examine, for example, the relative importance compared to the Earth origin ions.
1752 Ideally, we need new observations using dedicated instruments with much higher
1753 sensitivity for high mass (cf. Sect. 5). This is particularly important for the magnetospheric
1754 metallic ions because the ionospheric or mesospheric can also supply a significant amount
1755 and easily hide the lunar signal without specifying the external condition. However, the
1756 total Moon-origin ion flux might not be negligible when the Moon is upstream and the
1757 geomagnetic activity is moderate or quiet.

1758

1759 **4.1.3 Low-energy ions near the Moon** (Kaguya or Change-4, combining with ACE)

1760 For the source environment near the Moon, it is again useful to compare the lunar ion flux
1761 leaving the Moon with the solar wind. With the capability of detecting these ions, Kaguya
1762 observed notable increase of the Moon-origin ions around the Moon when CIRs arrived at
1763 the Moon, as mentioned in Sect 2.4.3. Unfortunately, the operation of Kaguya/IMA was
1764 only from 2008 to early 2009, i.e., during the lowest solar activity in the space age
1765 (deepest minimum), and only very few CIRs of minor intensity occurred. This is not easy
1766 because such a study requires a good constellation of Kaguya (the orbit plane with respect
1767 to the Moon-Earth line changes depending on the Earth's season), a magnetospheric
1768 satellite (in the magnetosphere and lunar wake) and the Moon (e.g., at few days before
1769 and after new Moons) during the CIR arrival. Probably for these reasons, we could not find

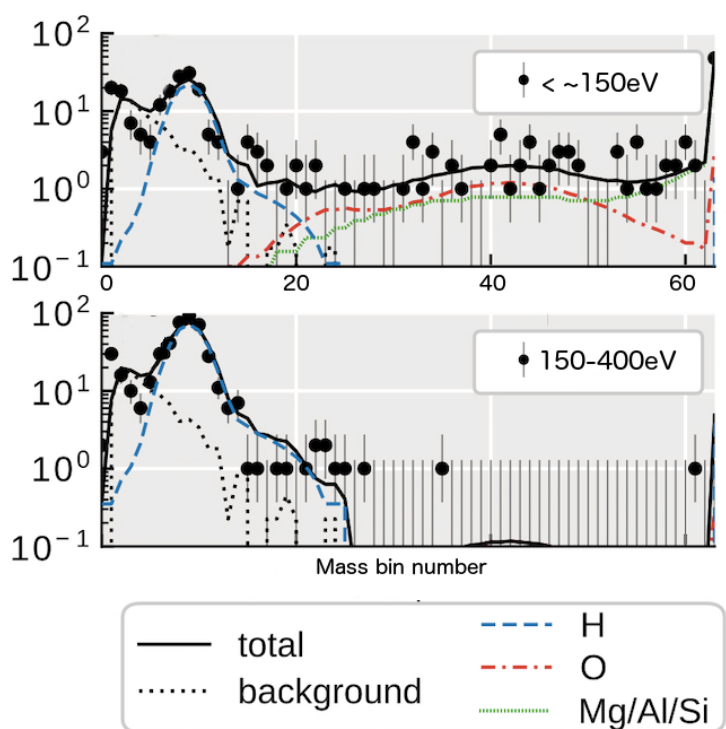
1770 a notable correlation between the Kaguya's soil-origin ion counts (time resolution of 1 day)
 1771 and Cluster CODIF data.

1772

1773 Other than CMEs or CIRs, the Moon is exposed to high upstream plasma energy in the
 1774 Earth's plasma lobe and mantle region because of the high flux of terrestrial escaping O^+
 1775 (Slapak et al., 2017; Yamauchi, 2019) when the Moon is in the magnetotail for about 5
 1776 days every month. For the Earth's plasma sheet, the Moon is exposed to hot (high
 1777 energy) ions including terrestrial O^+ , but the Kaguya observations for such periods are too
 1778 short for the sensitivity of IMA instrument to examine the expected increase of sputtering
 1779 inside the plasma sheet.

1780

1781



1782

1783 Figure 4.3: <<Asan2020egu2>> Energy-integrated mass spectrum of neutrals coming from
 1784 the lunar surface observed by Chang'e4/ASAN during January - September 2019. The
 1785 colored curves are modelled values assuming specific species. Upper: energy <math>< 150\text{ eV}</math> is
 1786 integrated. Lower: energy $> 150\text{-}400\text{ eV}$ are integrated. Comparing them, the high mass
 1787 signals for <math>< 150\text{ eV}</math> indicates the heavy species.

1788

1789

1790 Finally, the Chang'e-4 lunar rover is equipped with an ion/neutral instrument, the
 1791 Advanced Small Analyzer for Neutrals: ASAN (Wieser et al., 2020a). ASAN is capable of
 1792 separating the heavy species from proton/hydrogen (Wieser et al., 2020b), and is still

1793 operating after 5 years on the Moon surface since 2019. Including the FOV toward the
1794 lunar surface, ASAN can distinguish between the sputtering component and reflecting
1795 component based on energy analysis of the registered neutrals (Xie et al. 2021). Although
1796 its operation in many short (10-20 min) sessions give a total of only several hours every
1797 month, the much longer duration of the mission covers the inclining phase of solar cycle
1798 25, and has already experienced several strong CMEs such as the one in December
1799 2020. The count rate is marginally sufficient to separate H^+ and other elements (O^+ and
1800 heavier together), but it is still useful to examine the increase of sputtering heavy ions by
1801 the CME passage. Analysis is on-going.

1802

1803

1804

1805 **4.2 Ionospheric origin of low-energy heavy molecular ions (in-situ observations)**

1806

1807 **4.2.1. Recent observations above the ionosphere**

1808 With the ability to measure low-energy ions up to ~ 100 eV, which is sufficient for
1809 measuring most ion upflow and outflow events in the full altitude range of the CASSIOPE
1810 (Swarm-E) satellite (perigee 325 km and initial apogee 1500 km), the e-POP/IRM
1811 instrument is capable of separating the heavy molecular ions from the N^+ and O^+ (Yau et
1812 al., 2015, 2016) in the topside ionosphere. It covers both the high-latitude (around the
1813 dayside cusp) outflow region and the auroral and sub-auroral outflow region. In a
1814 preliminary study (Foss, 2019; Foss et al., to be submitted 2024), upflow of the heavy
1815 molecular ion above the ionosphere are often detected on the equatorward side of the
1816 auroral ion outflow region, especially during geomagnetic active periods, in addition to in
1817 the dayside polar cap.

1818

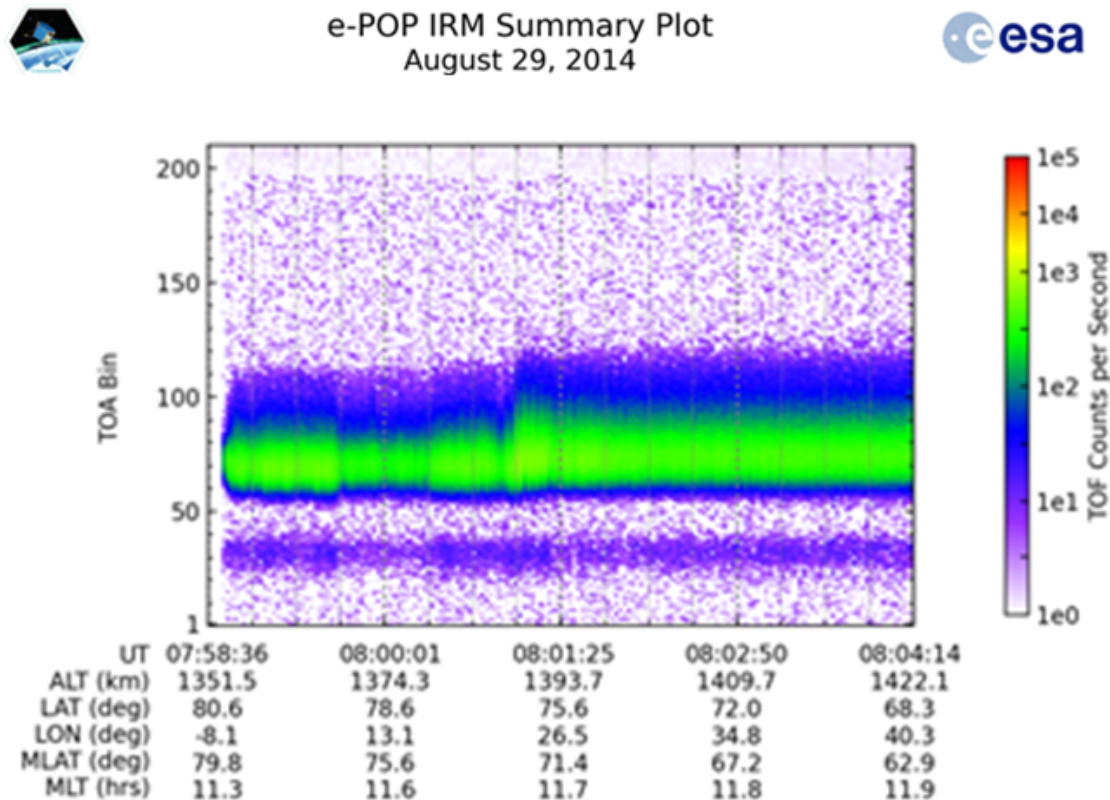
1819 Figure 4.4 shows one example of the e-POP/IRM observation in the dayside at ~ 1400 km
1820 altitude when $K_p = 3$ and $Dst = -37$ nT during the recovery phase of a moderate
1821 geomagnetic storm on 29 August 2014 (Dst minimum - 63 nT). The vertical axis is the
1822 time of arrival (TOA) at the detector which represents the TOF, and molecular ions
1823 correspond to TOA ~ 80 -100. Both the count and TOA increased during 08:00:55 -
1824 08:01:25 UT, at which the spacecraft is in the dayside at around $\sim 72^\circ$ magnetic latitude

1825 and ~ 1400 km altitude. This means that the molecular ions appeared in addition to O⁺.
 1826 Thus, e-POP can separate the heavy molecular ions from the atomic ions.

1827

1828 Figure 4.5 shows another example of the e-POP/IRM observation in the nightside at the
 1829 altitude of 1360 km when Kp = 6+ and Dst = -87 nT during a major geomagnetic storm
 1830 (Dst minimum -130 nT). Both the count and TOA suddenly increased at around 16:15:30
 1831 UT in the evening sub-auroral latitude (20.4 MLT, 53° Mlat.) indicating a mixture of
 1832 molecular ions and atomic ions, with continued high count in the TOA range corresponding
 1833 to atomic ions. This example shows that, unlike Akebono observation at higher altitude,
 1834 heavy molecular ions can reach the topside ionosphere in the nightside sub-auroral
 1835 region, at the equatorward side of the atomic ion outflow region in the auroral region.

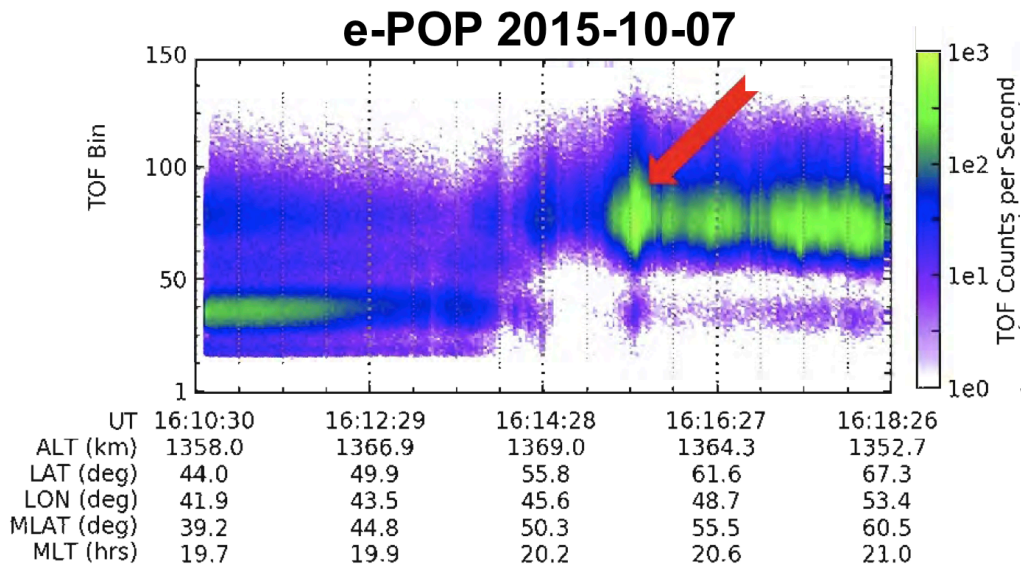
1836



1837

1838 Figure 4.4 <ePOP-MI_sample_cut>: TOF-time spectrogram of observed ion count rates on
 1839 e-POP during the recovery phase of a magnetic storm on 29 August 2014. Vertical axis is
 1840 time of arrival at detector (TOA) which represents the TOF, and molecular ions correspond
 1841 to TOA ~ 80-100.

1842



1843

1844 Figure 4.5 <ePOP_2015_1007>: Same as Figure 4.4 but for on 7 October 2015. This
 1845 traversal took place just before the lidar event in Figure 4.12a.

1846

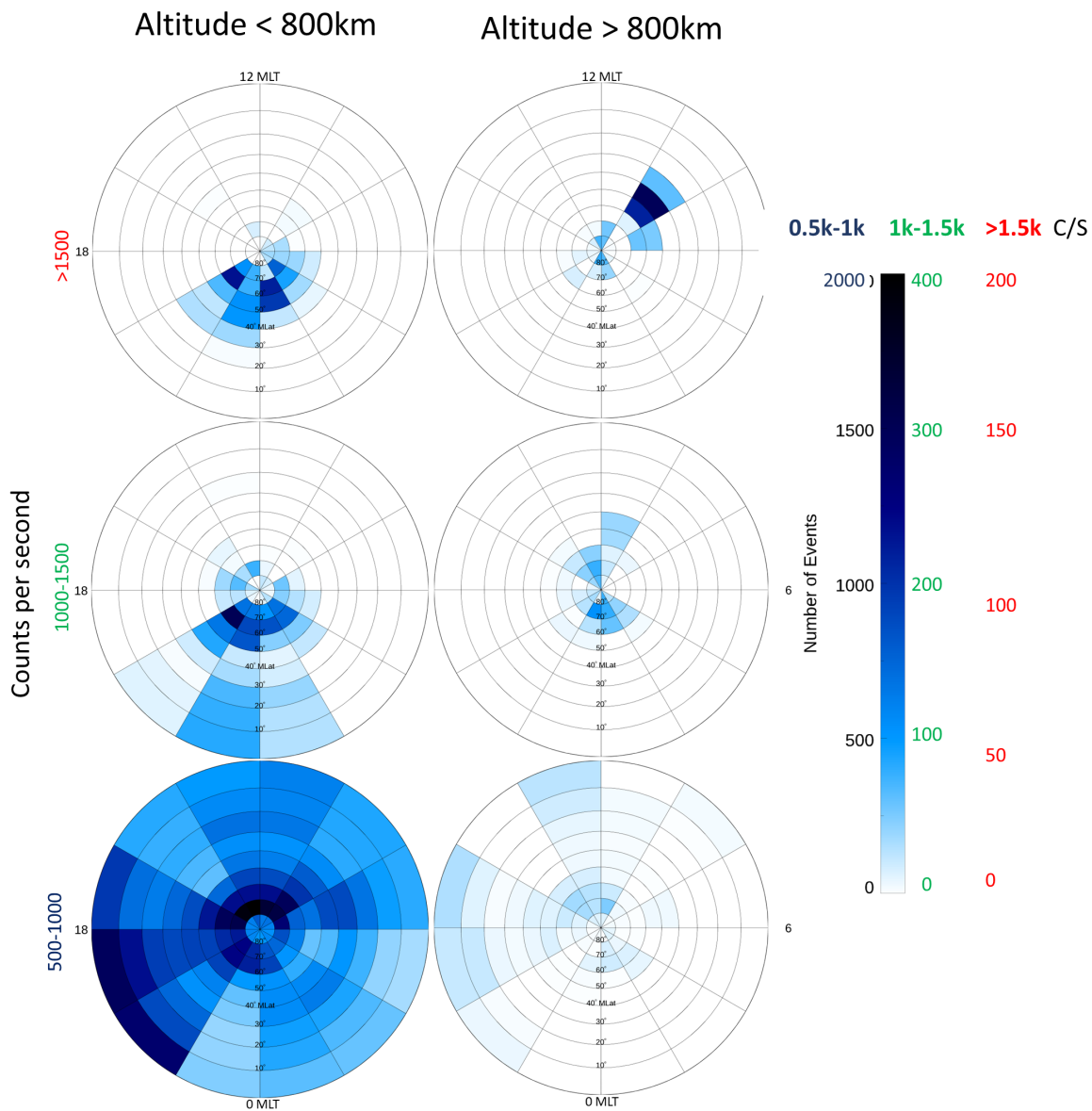
1847

1848 The observed low- and mid-latitude events are predominantly low- or medium-flux events
 1849 at low altitudes (<800 km). We here note that the lowest-latitude (<10° MLAT) events at
 1850 16-22 MLT are believed to be mostly associated with the Appleton anomaly and not part of
 1851 the molecular ion upflow population. Otherwise, the other low-latitude detection of the
 1852 molecular ions (particularly for < 800 km altitude) suggests that these molecular ions are
 1853 convected equatorward in the ionosphere after originating at higher (auroral) latitudes.
 1854 Thus, the ionospheric convection (both equatorward and upward) can be strong enough to
 1855 transport heavy molecular ions to very low latitudes before they reach the topside
 1856 ionosphere or before they undergo dissociative recombination to form a pair of neutral
 1857 atoms (see Sect. 2.2.2).

1858

1859 The detection of molecular ions on the nightside by e-POP at 1400 km answers one of the
 1860 questions in Sect. 2.2: at which altitude do the molecular ion upflows begin to disappear in
 1861 the nightside outflow region where atomic ion upflows are detected at much higher
 1862 altitudes such as Akebono and Polar? The difference in the observed ion upflows between
 1863 molecular ions and atomic ions starts at about 1200 km altitude on the nightside. There
 1864 are two possibilities for the cause of such difference between the molecular ions and
 1865 atomic ions: insufficient pre-energization to reach the altitude at which the transverse ion

1866 energization starts (above 2000 km; Whalen et al., 1990; 1991), and the absence of main
 1867 energization at the latitude of molecular ion formation and upflow.
 1868



1869
 1870 Figure 4.6: Statistical distributions of molecular ion detection observed by e-POP as a
 1871 function of MLT and magnetic latitude (MLAT) below (left) and above (right) 800
 1872 altitude, respectively; top to bottom: peak count rate >1500, = 1000-1500,
 1873 and = 500-1000 counts-per-second, respectively. (Courtesy of V. Foss, to be submitted 2024)
 1874

1875
 1876 For the first scenario, the pre-energization that is required above e-POP altitude for a
 1877 molecular ion to reach 4000 km (2500 km) altitude must be less than 4 eV (2 eV) in the
 1878 field-aligned direction, which is double (about the same as) the energy required from 500
 1879 km altitude to initial e-POP apogee (1500 km altitude) according to Table 2.1 in Sect. 2.

1880 For the second scenario, energization of ions between the e-POP altitude and Akebono
1881 altitude including energization of all types must be less than 7 eV in the field-aligned
1882 direction. Neither of these two possibilities can be examined due to the lack of dedicated
1883 instruments for observing molecular ions in the magnetosphere.

1884

1885

1886 **4.2.2. Magnetospheric molecular ion measurements by non-optimized instruments**

1887 There are instruments that are not optimized for separating molecular ions from atomic
1888 ions, but still able to identify them under certain conditions, particularly if the ion outflow is
1889 very strong. We here show an example from the Cluster CODIF instrument.

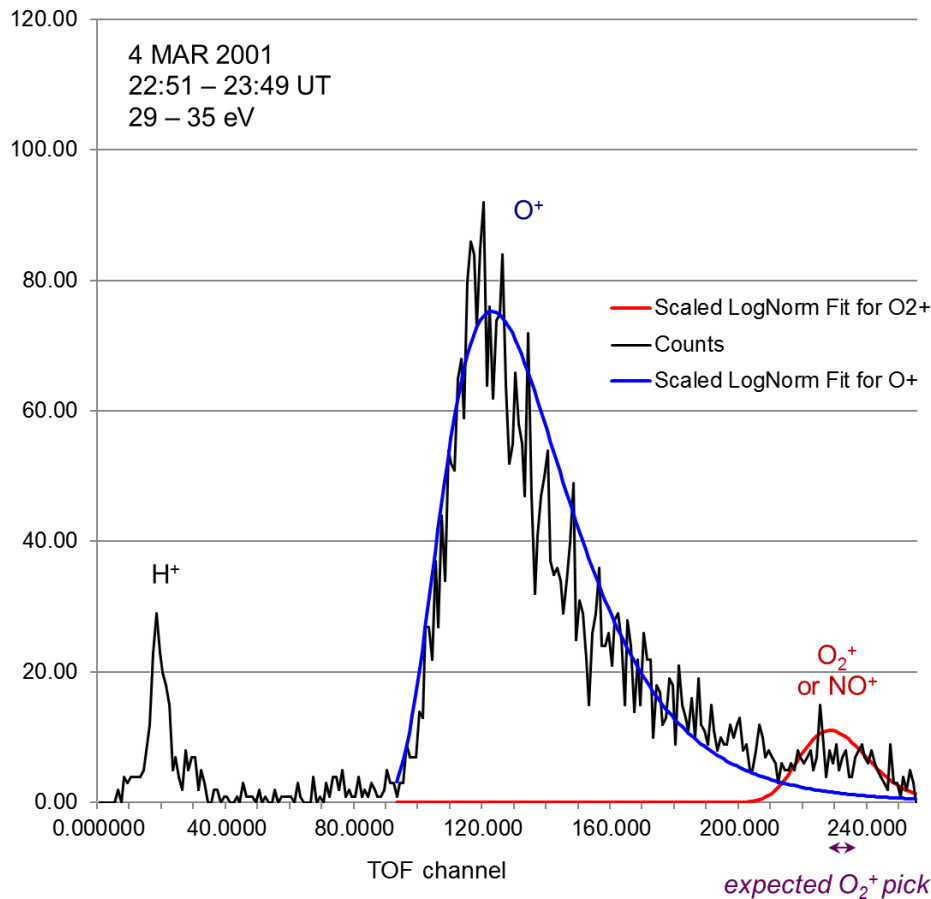
1890

1891 The CODIF ion mass spectrometer onboard Cluster, with a mass resolution of $m/\Delta m \approx 5-7$
1892 (Rème et al., 2001), was not designed to separate molecular NO-group ions and atomic
1893 CNO-group ions. Ions lose energy when going through the thin carbon foil in the
1894 instrument at the start of the time-of-flight section. This energy straggling in the foil leads to
1895 a long tail in the time-of-flight distribution of heavy ions. Therefore, the time-of-flight peak
1896 of the heavy molecular ions on CODIF overlaps with the tail of the O⁺ peak, making it
1897 difficult to separate the molecular ions. Usually, the molecular ion counts are negligible
1898 compared to the O⁺ tail. Therefore, the molecular ion counts have not been counted.

1899

1900 However, during some strong events, the flux of the molecular ions occasionally becomes
1901 high enough to have a separate peak above the oxygen tail in the TOF region where the
1902 molecular ion peak should appear (Dandouras et al., in preparation, 2024), as shown in
1903 Figure 4.7. In such cases, the molecular ions can be distinguished. By fitting multiple
1904 peaks to the time-of-flight spectra, the abundance of molecular ions can also be estimated.

1905



1906

1907 Figure 4.7 <lannis_Figure 4a_CIS TOF fit> : Time-of-flight (TOF) histogram for the 29 – 35
 1908 eV ions detected by CODIF onboard Cluster on 4 March 2001 between 22:51 and 23:49
 1909 UT above the southern polar cap, while observing an upwelling ion beam. The blue and
 1910 red curves represent the simulation results for atomic O⁺ (blue curve) and fragments of
 1911 molecular O₂⁺ (red curve) ions entering the TOF section of the instrument, after their
 1912 passage through the carbon foil where the molecular ions fragment (SRIM software
 1913 simulation). The upwelling ion beam is dominated by the O⁺ ions, but the instrument TOF
 1914 data also show the existence of a weak O₂⁺ (or NO⁺) population.

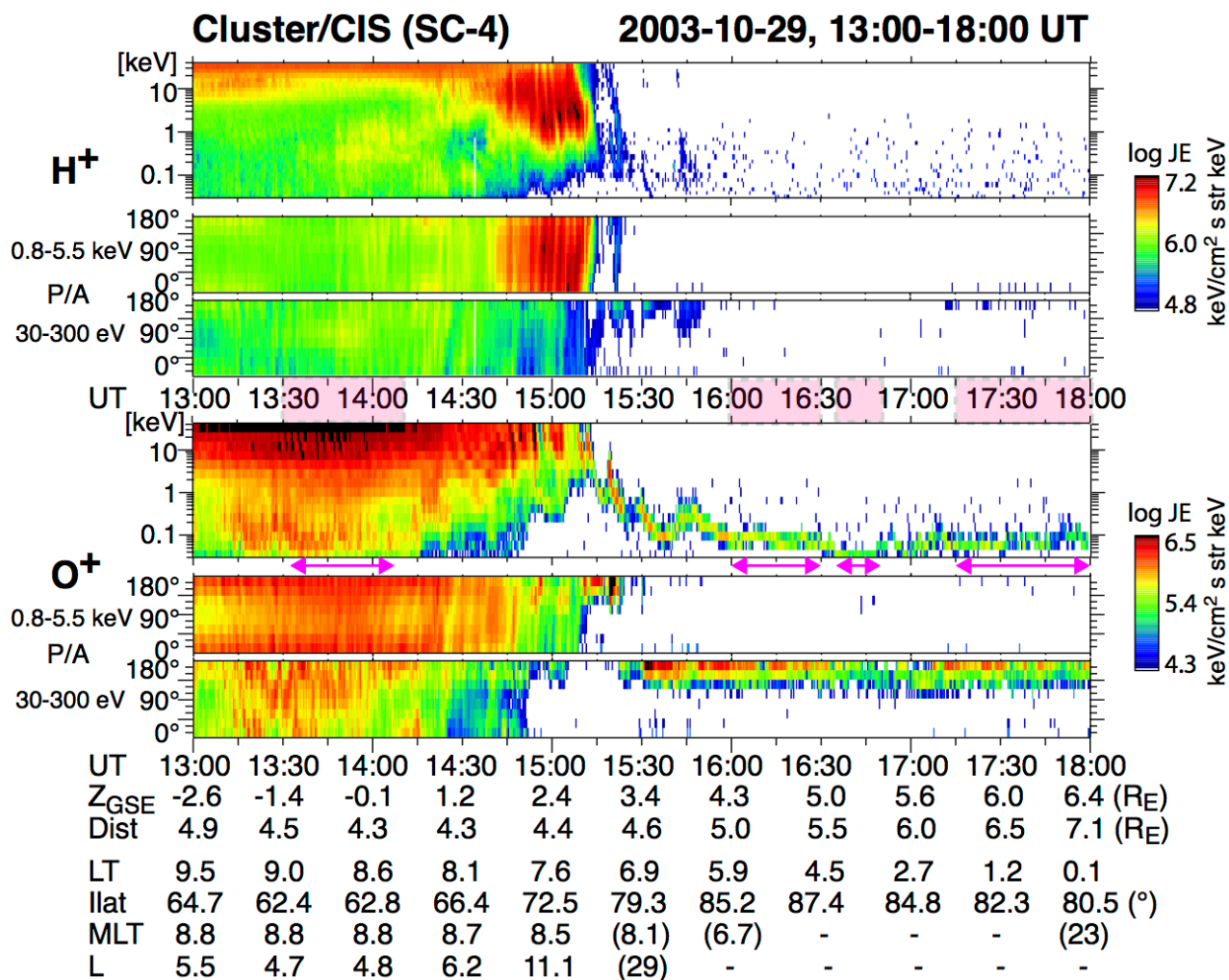
1915

1916

1917 Figure 4.8 shows one Cluster pass through the inner magnetosphere and over the polar
 1918 cap during the 2003 Halloween storm. The top three panels show the H⁺ energy spectrum
 1919 and the pitch angle distributions for two energy ranges. The next three panels show the
 1920 same parameters for O⁺. Two clear spatial regions can be distinguished. From the
 1921 beginning of the plot until ~15:15, Cluster is in the closed field line region of the inner
 1922 magnetosphere. After 15:15 UT, it is moving out over the polar cap, and observing a
 1923 narrow distribution of O⁺ ions that is convecting tailward, likely from the cusp region.

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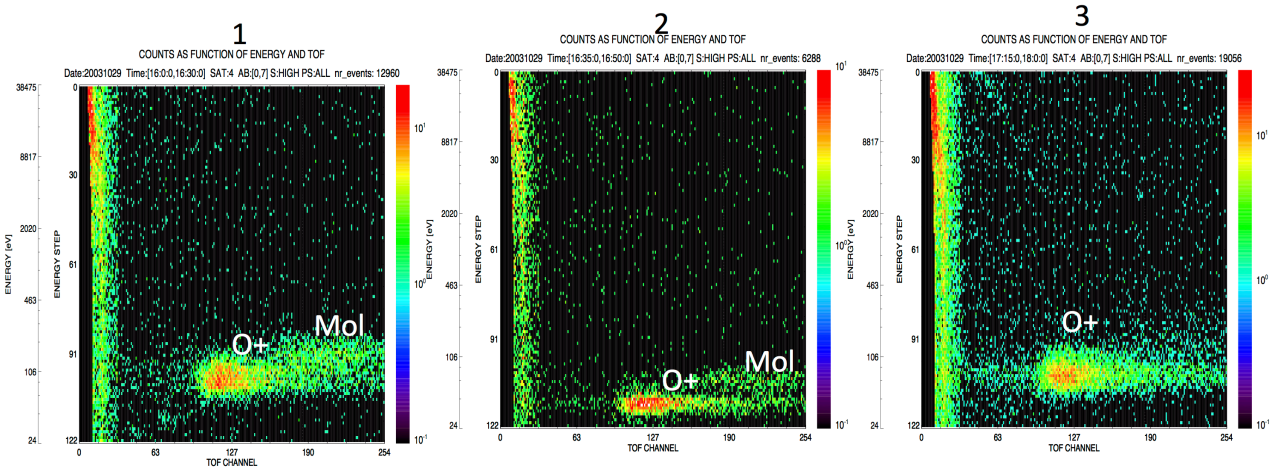
1941

1942

1943

Figure 4.8 <HO_20031029b>: Cluster CIS/CODIF observation of Halloween storm on 29 October 2003. Energy-time and pitch angle-time spectrograms for H⁺ (upper panels) and for heavy ions (labeled as O⁺) are displayed. The periods indicated by horizontal arrows and hatches correspond to periods when the time-of-flight distributions were obtained, as shown in Figure 4.8 (16:00-16:30 UT, 16:35-16:50 UT, and 17:15-18:00 UT) and Figure 10 (13:30-14:10 UT), respectively.

For ions flowing over the polar cap where solar wind electric field drives ion convection perpendicular to the magnetic field, the combination of the field-aligned upflow and this perpendicular convection leads to the “velocity filter effect”, in which the ions become spatially separated by their velocity. In Figure 4.8, such a narrow energy-banded O⁺ is seen after 15:10 UT. The pitch angles of these O⁺ are nearly against the magnetic field, i.e., flowing outward from the Earth's northern hemisphere. Such a spectrogram with narrow band O⁺ is typically seen in the polar cap.



1944

1945 **Figure 4.9** <cluster_O2_20031029>: Energy versus time-of-flight scatter plot of ion events
 1946 measured by Cluster CIS/CODIF during the Halloween storm on 29 October 2003. (a)
 1947 16:00-16:30 UT, (b) 16:35-16:50 UT, and (c) 17:15-18:00 UT. These periods are indicated
 1948 by horizontal arrows and hatches in Figure 4.8. Horizontal axis is time-of-flight (TOF),
 1949 which is inversely proportional to the velocity in the instrument. The vertical axis is ion
 1950 energy (channel 80: ~500 eV; channel 90: ~200 eV; and channel 105: ~90 eV).
 1951

1952

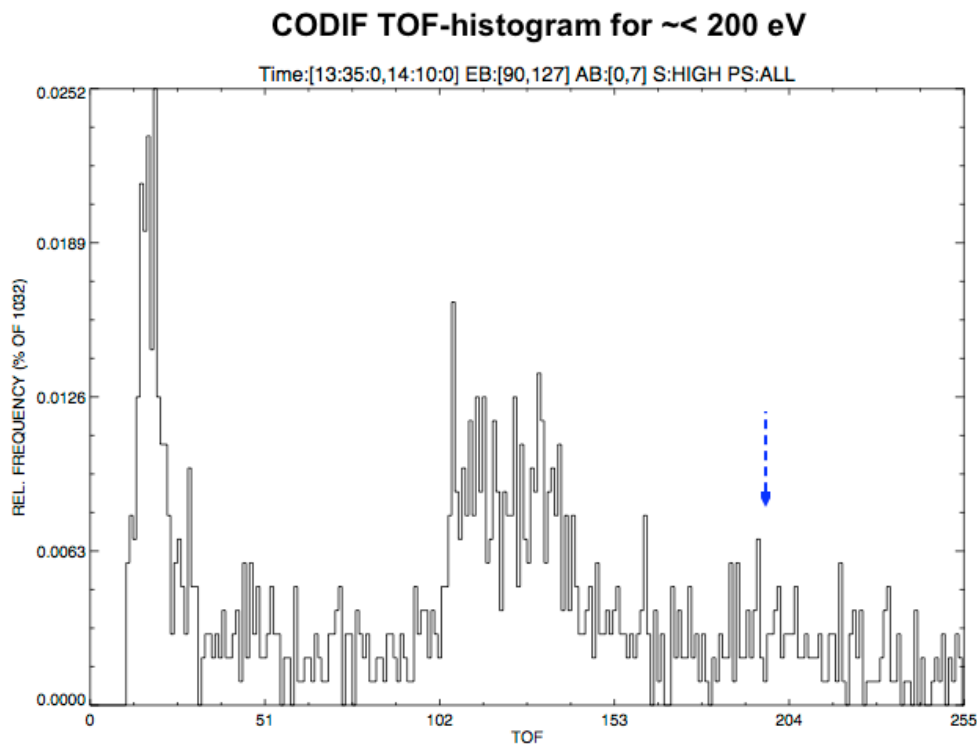
1953

1954 For this time period (after 15:10 UT), the time-of-flight (TOF) spectra of the CODIF data
 1955 can be used to determine if molecular ions coexist with O⁺. Figure 4.9 shows a scatter plot
 1956 of energy versus TOF for three time periods labeled in Figure 4.8. During the first two time
 1957 periods (16:00-16:30 UT and 16:35-16:50 UT) both in the morning sector in the polar cap,
 1958 the low-energy O⁺ ions are evident, with a peak at a TOF channel around 100, and a long
 1959 tail towards higher TOF. But the O⁺ peak is accompanied by a second track at higher
 1960 energy and longer time-of-flight consistent with molecular ions. Such separation in energy
 1961 of the outflowing ions is typically seen in the polar cap between H⁺ and O⁺ (e.g., Nilsson et
 1962 al., 2006), and hence is attributed to mass difference for Figure 4.9, too. During the last
 1963 time period (17:15-18:00 UT, the midnight sector near the polar cap boundary), only the
 1964 O⁺ track is observed showing that the molecules from the dayside outflow are not reaching
 1965 this location.

1966

1967 There are also low energy field-aligned ions observed during the inner magnetosphere
 1968 perigee pass ($L < 5$). The pitch angles of O⁺ at 30-300 eV, shown in the bottom panel of
 1969 Figure 4.8, have peaks close to either at 0 or 180 degrees. The field-aligned dominance
 1970 indicates that these heavy ions directly come from the ionosphere, including those trapped
 1971 and bouncing after outflowing from even lower latitude (Quinn and McIlwain, 1979). In
 both cases, the source is at lower latitude than the ions seen in the polar cap (e.g., after

1972 15:10 UT in Figure 4.8). To evaluate the existence of molecular ions, we again examine
 1973 the time-of-flight. Figure 4.10 shows the result for the ions at 30-200 eV during 13:15 -
 1974 14:10 UT (perigee pass at $L < 5$) where the low-energy field-aligned O^+ is observed. Here
 1975 we show a histogram instead of a scatter plot. The expected TOF location of the molecular
 1976 ions is marked with the blue arrow. A clear enhancement is observed there, indicating that
 1977 molecular ions were outflowing not just in the cusp region, but in the lower latitude closed-
 1978 field line region at $< 62\text{-}63^\circ$ Inv, as well, for this intense storm.
 1979



1980

1981 **Figure 4.10** <fromPPT_Lynn1330_1410>: Time-of-flight Histogram of ion events in the
 1982 energy range 30-200 eV during 13:15 - 14:10 UT, where outflowing ions are observed.
 1983 The vertical dashed arrow corresponds to the expected location of molecular ions (m/q ~
 1984 30).
 1985

1986

1987 4.2.3 Ground-based data

1988 To investigate the ion upflow, incoherent scatter (IS) radars such as EISCAT VHF and
 1989 UHF radars have long been used to infer the extraordinary upward convection up to about
 1990 500 km altitude (e.g., Wahlund et al., 1992; Ogawa et al., 2019, Takada et al., 2021).
 1991 However, these IS radar observations cannot separate the ion composition, and need
 1992 assumptions to estimate the heavy ion upflow (assume certain composition ratio of H^+ , O^+ ,

1993 and N_2^+ for EISCAT case). Furthermore, for the actual ionospheric observations, the
1994 obtained maximum upward ion velocities are a few hundred m/s ($\sim 10^{-3}$ eV), which are
1995 negligible compared to the escape velocity (10^1 eV). Therefore, the upflow observation by
1996 these IS radars does not necessarily mean an upflow above the exobase, or, more
1997 importantly, must not be interpreted simply as the upflow of molecular ions at higher
1998 altitudes, as shown in Sect 4.2.1 (significant high-altitude/low-altitude difference observed
1999 by e-POP and Akebono). The heavy ion upflow at sub-auroral latitudes (where e-POP
2000 detected molecular ions with sufficient flux) needs sufficient additional energization (via
2001 wave-particle interactions, for example) to the magnetosphere, and this is not easy
2002 because of the lower electromagnetic activity than the auroral latitude.

2003

2004 Thus, using an IS radar to estimate the molecular ion motion is very misleading and is not
2005 recommended unless one makes a statistical study with the radar data and e-POP
2006 observation using many traversals. On the other hand, the EISCAT radars can monitor the
2007 general upflow condition, allowing us to define the necessary condition for molecular ions
2008 to reach the topside ionosphere, particularly for the dayside polar cap source.

2009

2010

2011

2012 **4.3 Upper atmospheric source of metallic ions** (Ground-based observation, Model)

2013

2014 **4.3.1 Re-visit of Fe⁺**

2015 As covered in Sect. 2.3, there are layers of neutral metal atoms in the atmosphere that
2016 appear globally between about 75 and 110 km as well as layers of ionized metal atoms
2017 between about 90 and 130 km (Plane 2003; Plane et al., 2015a). As discussed in Sect.
2018 2.3.3, these metallic ions in the lower part of the ionosphere can reach the exobase
2019 ionosphere where heavy molecular ions are detected because of the similar mass as
2020 these molecular ions. The same mass-argument applies to the energization process above
2021 the ionosphere; i.e., metallic ions that have arrived at the exobase should reach the
2022 magnetosphere. However, with a mass only twice the O₂⁺ mass, Fe⁺ (mass 56) was
2023 detected with only 350 counts by Geotail over 20 years (about 2 count per month), which
2024 is far below the detection of the heavy molecular ions in the magnetosphere (Sect 2.2).
2025 This low detection rate can set the boundary condition for ionospheric transport models for
2026 vertical ion transport, which are still under development (e.g., Shinagawa and Oyama,
2027 2006).

2028

2029

2030 **4.3.2. Lidar observation combining with magnetospheric patchy data**

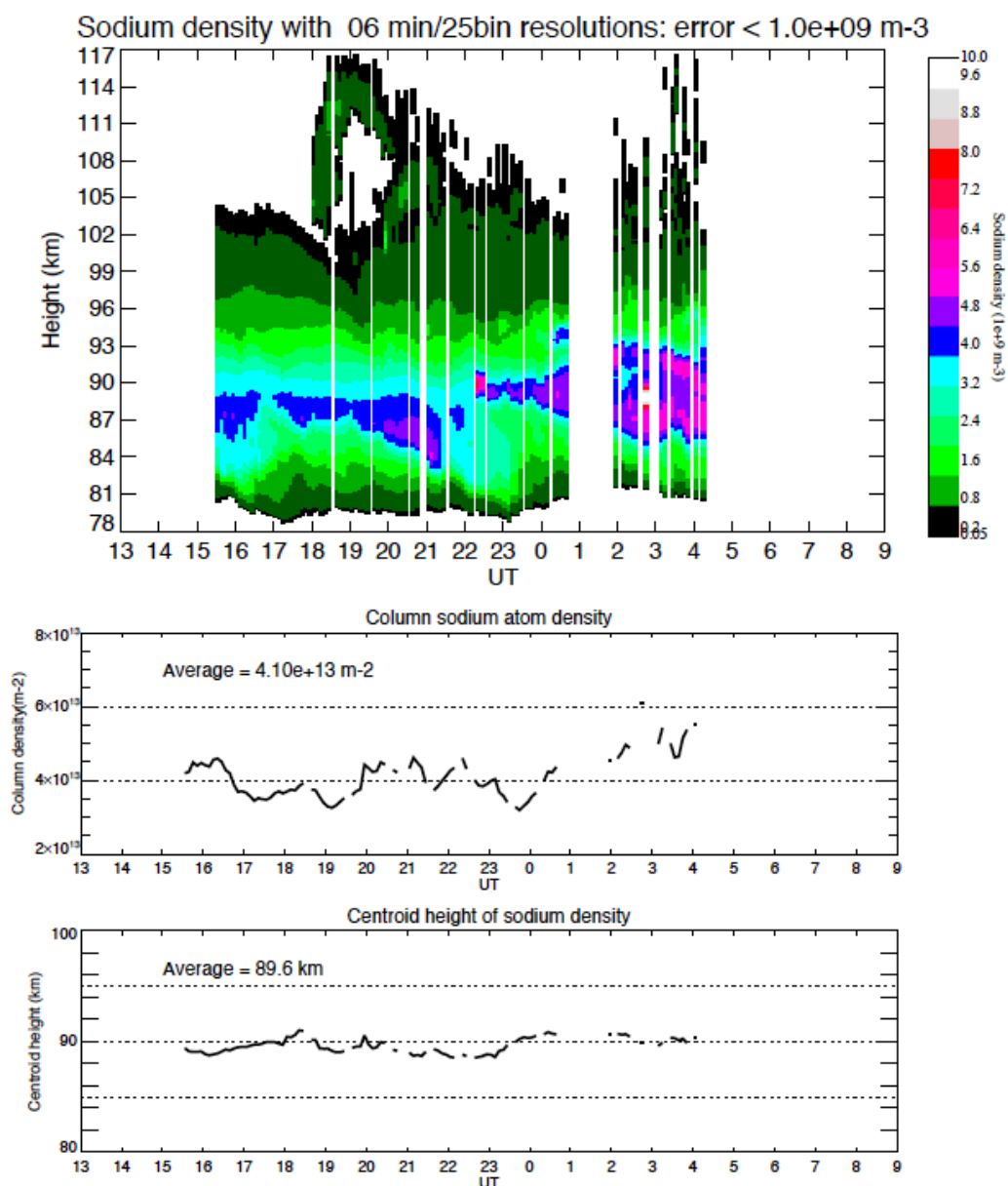
2031 As summarized in Sect 2.3.2, metallic ions (Na, K, Fe, Ca, Ca⁺, Li, Ni and AlO) in the lower
2032 ionosphere and thermosphere are mainly observed by many methods. If limited to low
2033 altitudes below the mesosphere, the modern resonance lidar is able to make diurnal
2034 observations (Plane, 2003). Even the vertical velocity can be measured (e.g., down to 1
2035 cm/s resolution for Na atoms) between 80 and 105 km by Na lidar (Gardner et al., 2014).
2036 The vertical fluxes for other species can also be estimated by co-locating the other lidar
2037 (e.g., Fe lidar).

2038

2039 Although the altitude range for the lidar measurement covers only the lower part of the
2040 ionosphere, such monitoring helps qualitatively understanding the dynamics of metallic
2041 species during the ionospheric conditions that enhance the molecular ion upflow above the
2042 exobase because the required upward convection for such transport is common (“strong”)
2043 for both the metallic ions and heavy molecular ions, as mentioned in Sect. 2.3.3. In this
2044 sense, monitoring the dynamics of metal layers (AlO, Ni, Na, and Fe with existing
2045 technology) in the ionosphere when the molecular ion upflow is observed by low-altitude

2046 satellite could be useful because any lidar data can potentially be compared with low-
 2047 altitude satellite observations that is capable of separating heavy ion (mass > 20 amu)
 2048 such as e-POP. For example, statistics of conjugate observations for better understanding
 2049 of the dynamics of the metal species in the condition that molecular ion upflow is
 2050 enhanced.
 2051

Variation of sodium density on November 1, 2012



2052
 2053 Figure 4.11 <nozawa_lidar_121101>: Sodium (Na) atom density (top), column sodium
 2054 atom density (middle), and the centroid height (bottom) with 6 min/1 km resolutions
 2055 obtained with the sodium LIDAR at Tromsø on 1 November 2012. This particular sodium
 2056 lidar at Tromsø cannot make observations under sunlit conditions (this is why the
 2057 observation started late).

2058

2059

2060 Here we show an example for Na atom observation. The sodium atom layer usually exists
2061 between 80 and 110 km with a peak height between ~87-92 km. The lower edge of the
2062 sodium layer is sharp with a scale height of 2 – 3 km, reflecting the rapid conversion of Na
2063 into molecules such as NaHCO₃ (Plane, 2003). The top-side of the layer also has a small
2064 scale-height because of the conversion of Na into Na⁺ in the lower ionosphere. Figure 4.11
2065 shows sodium atom density data obtained at Tromsø (67° geomagnetic latitude; (69.6°N,
2066 19.2°E) in geographic coordinate) (Nozawa, et al., 2014) over 11 hours, starting from
2067 15:30 UT on 1 November 2012.

2068

2069 On this particular night (1 November 2012), the upper part of the sodium layer extended
2070 above 110 km from about 18 to 20 UT. The column sodium density (middle panel) varies
2071 with time, and does not show any particular increase during the time interval. The centroid
2072 height of the sodium density (bottom panel) varies with time in height range between 89
2073 and 91 km, and again no special feature is found during 18 and 20 UT. It was rather
2074 geomagnetically active interval with 3 hourly local K index (3-hour resolutions) at Tromsø
2075 being 6 (15-18 UT) and 6 (18-21 UT). Thus, although the cause of the extension of the
2076 sodium density above 110 km is not clear at a moment, auroral activity could be involved.

2077

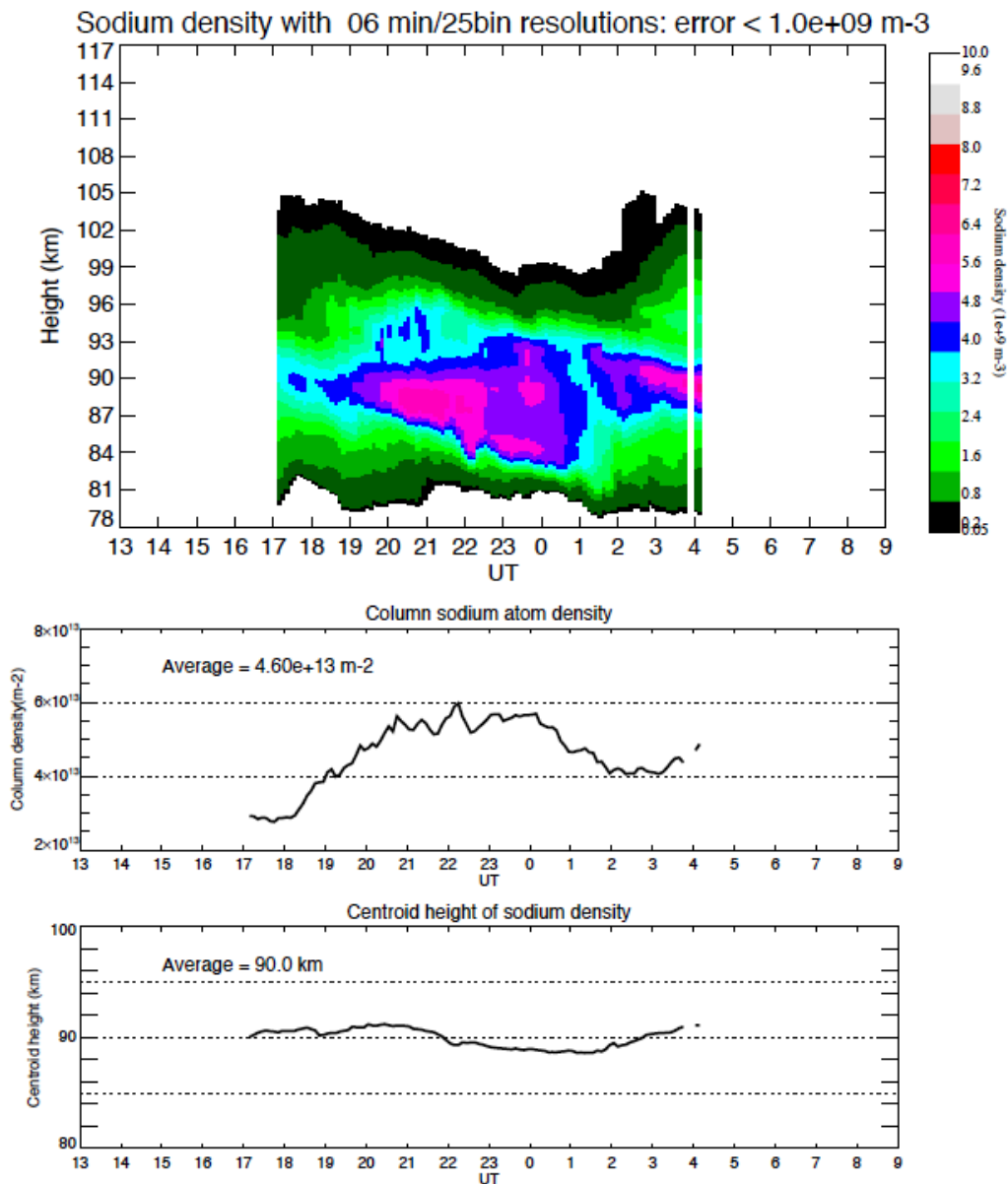
2078 Figure 4.12 shows one such example on 7 October 2015 during the major storm as
2079 summarized in Sect. 4.2.1. Tromsø Na lidar data (sodium density) started observation
2080 from 17 UT, about the one hour after e-POP detected heavy molecular ion upflow at
2081 around 20.4 MLT (corresponds to about 19 UT at Tromsø) as shown in Figure 4.6. The
2082 local K-index (3-hour resolutions) at Tromsø was 5, 7, 6, 4, 5 from 15 UT on 7 October
2083 2015 and AL reached < -1500 nT at 18-19 UT. While the e-POP observation for the
2084 traversals at later hours are not optimum, both Dst and AE are developing during the
2085 following few hours after the e-POP detection of molecular ions at around 16:15 UT.

2086

2087 During that few hours, the lidar detected significant variation of sodium density and
2088 thickness of the layer. The upper edge of the sodium layer (the density of $5 \times 10^7 \text{ m}^{-3}$) was
2089 located at 100 km at 17 UT, decreasing with time to reach about 95 km at 23 UT on the
2090 same night. The auroral activity was high, implying that the sodium density variation and
2091 the altitude variation can be partly due to the auroral effect and gravity waves,

2092 superimposed on the diurnal tidal variation. This upward expansion of the Na signal,
 2093 which is not as clear as the Figure 4.11 (1 November 2012), can be due to daily variation
 2094 but may also have some relation with enhanced access of the molecular ions to the
 2095 topside ionosphere, which is suggested from the e-POP observation.
 2096

Variation of sodium density on October 7, 2015



2097
 2098 Figure 4.12 <nozawa_lidar_151007>: Same as Figure 4.11a but for on 7 October 2015,
 2099 right after the e-POP event in Figure 4.12b.
 2100

2101
 2102 Finally, very long-time monitoring of the metal layer by lidar potentially have another merit
 2103 from space safety monitoring viewpoint. For elements that the space debris has much

2104 more fraction than meteorite, we might be able to see the effect of ablation of space debris
 2105 (space waste) as the new deposit.

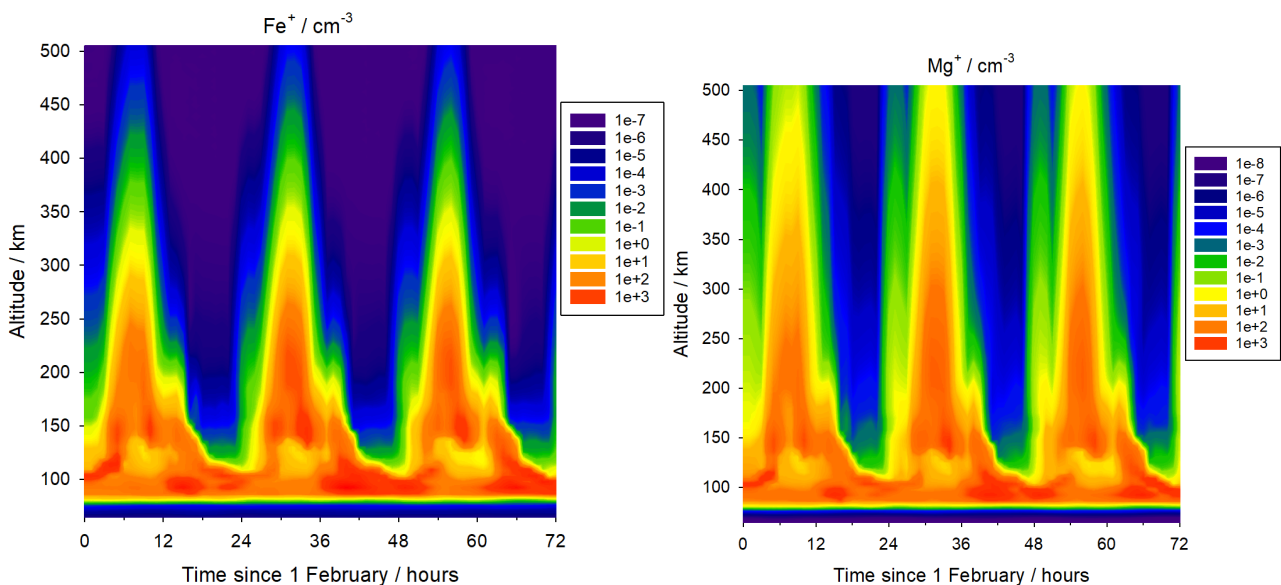
2106

2107

2108 4.3.3 Model of neutral wind for heavy elements

2109 The possibility of metallic ions reaching the exobase and magnetosphere can be explored
 2110 using models of the upper atmospheric transport of heavy elements to high altitudes
 2111 potentially beyond the exobase. Recently, two global circulation models, SAMI3 (Huba et
 2112 al., 2019) and WACCM-X (Wu et al., 2021), have been developed to study the transport of
 2113 metallic ions in the thermosphere. WACCM-X is a chemistry-climate model that includes
 2114 the injection of metals from meteoric ablation, and the full neutral and ion-molecule
 2115 chemistry of Fe ($m = 56$), Mg ($m = 24$) and Na ($m = 23$). This model shows that metal ions
 2116 are transported to altitudes above 400 km at low geomagnetic latitudes by $\mathbf{E} \times \mathbf{B}$ forcing -
 2117 the so-called daily “fountain effect” (Wu et al., 2021).

2118



2119

2120 Figure 4.13 Modelled Fe^+ (left-hand panel) and Mg^+ (right-hand panel) concentrations
 2121 (units: ion cm^{-3}) calculated by WACCM-X for 3 days from February 1st, at 80°S and 90°E .
 2122 The time axis refers to UT. Although the highest Fe^+ and Mg^+ concentrations are in the
 2123 main layers below 120 km, there is significant uplift of the metal ions to ~ 500 km (the
 2124 model top) starting around 03:00 UT (0900 LT).

2125

2126

2127 Figure 4.13 shows an example of WACCM-X model output for the southern polar region
 2128 during average geomagnetic conditions in February (i.e., slightly after the summer solstice

2129 and perihelion, when ionizing solar radiation maximizes). The model predicts that both Fe⁺
 2130 and Mg⁺ daily reach 500 km altitude, which is the top of the model and most likely above
 2131 the exobase, even during average geomagnetic condition. By reaching the exobase, these
 2132 metallic ions may access the space in the same manner as molecular ions. Figure 4.10b
 2133 shows that more Mg⁺ than Fe⁺ reaches this altitude, reflecting its lower mass. This
 2134 behaviour is only seen in the model for southern polar latitudes during summer. The
 2135 corresponding uplift in northern polar latitudes during summer is much smaller.
 2136 Considering the high-altitude location where molecular ions are known to reach high
 2137 altitudes (Sect. 2.2), those metallic ions that reach the exobase might even access the
 2138 high altitude, particularly for disturbed periods when we expect stronger upward
 2139 convection and hence these metallic ions may reach the magnetosphere.

2140

2141

2142 **4.4 Re-entering space debris as a heavy ion source: Outlook and unanswered** 2143 **questions**

2144

2145 Table 2.5: Orbiting artificial objects

	2019-01	2022-12	2023-12
orbiting satellite*1	~ 5000	~ 9780	~ 11500
debris on catalogue	~ 22300	~ 32400	~ 35110
total mass of orbiting objects	> 8400 t	> 10500 t	> 11500 t
∑ fragmentation events	> 500	> 630	> 640
number of debris (>10cm)	~ 36500 (model estimate 2021)		
number of debris (1-10cm)	~ 1 million (model estimate 2021)		
number of debris (0.1-1cm)	~ 130 million (model estimate 2021)		

2146

2147

2148

2149

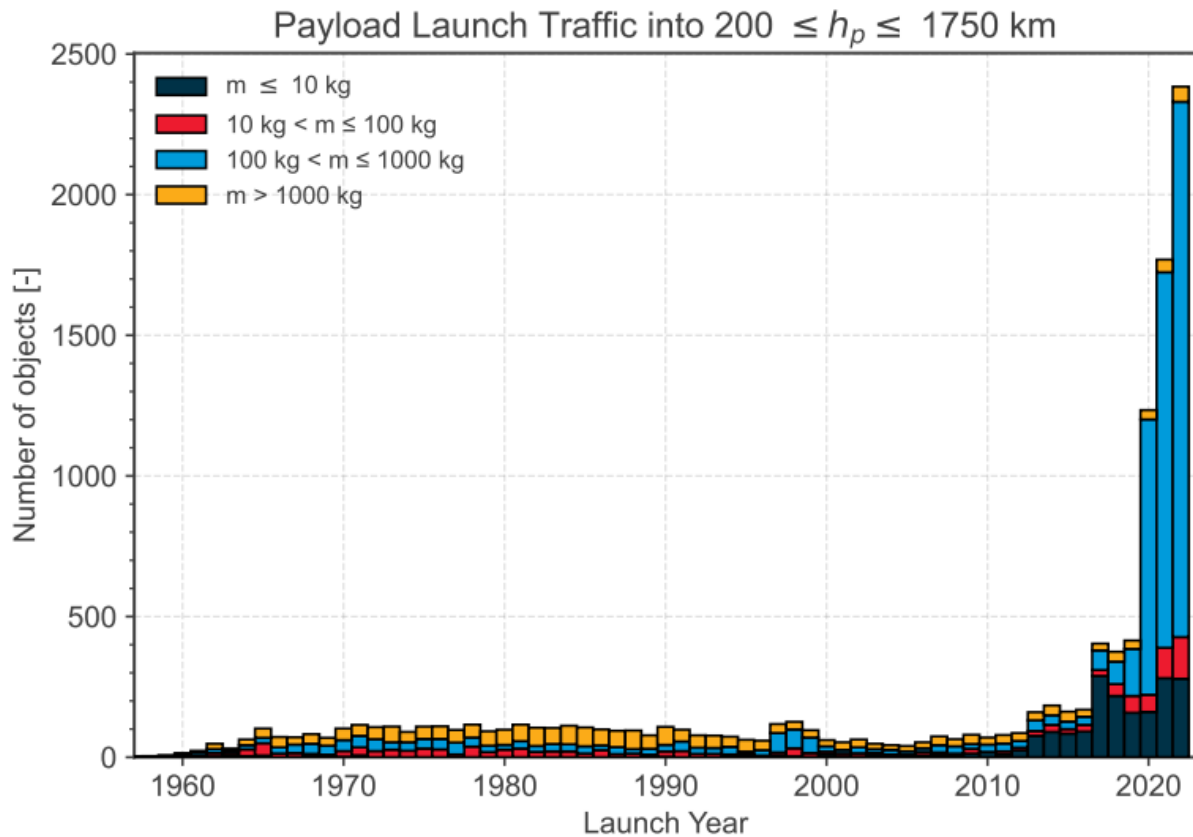
2150

2151 Today's society is heavily reliant on the space infrastructure (satellites), and one can
 2152 safely assume that this dependence will only increase in the near future (ESA, 2023a).

2153 The surge of large satellite constellations (LSC) consisting of several 1000 satellites due to
 2154 the commercialization of space will increase the number and mass of spacecraft launched
 2155 into Earth' orbit exponentially. The start of this exponential rise stemming from the
 2156 installation of LSCs can already be seen today, as depicted in Figure 4.14. Over less than

2157 five years, total mass of orbiting objects increased by 20%, their number by more than
 2158 50% as shown in Table 2.5.

2159



2160 Figure 4.14: ESA (2023a), Annual space environment report, September 2023
 2161 (www.sdo.esoc.esa.int/environment_report/Space_Environment_Report_latest.pdf)
 2162

2163

2164

2165 Decommissioning of spacecraft and remnants of launches lead to the re-entry of space
 2166 debris/waste into the atmosphere, where it ablates and injects atoms and ions as
 2167 described in Sect. 2.3.2. However, very few existing studies modelling spacecraft re-entry
 2168 focus on the atmospheric mass injection as this topic has been mostly disregarded in the
 2169 past.

2170

2171 Comprehensive atmosphere mass input calculations and observations of particles
 2172 transported downwards to stratosphere heights show that the injection of metal atoms and
 2173 ions into the atmosphere is already higher due to the ablation of space debris/waste than
 2174 the meteoroid input for some element species, currently Al, Cu, Ge, Li and Pb (Schulz et
 2175 al., 2021, Murphy 2023). Other rare metallic elements have also been observed (such as

2176 Nb, Ag, and Hf) and even traces of a large number of additional metals. In the future, the
2177 projected and already occurring strong increase in on-orbit spacecraft mass will lead to
2178 other element species injecting more mass compared to the natural input, namely Ti and
2179 Ni and a large number of the trace metals. Thus, the possibility of environmental effects
2180 and the contamination of the mesospheric heavy ion source should not be underestimated
2181 as even the rocket-boosters used to launch payloads into space contribute to the artificial
2182 influx as they partially ablate during re-entry (Schulz et al., 2021). It has even been
2183 suggested that the increase of re-entering material due to mega-constellations can begin
2184 an uncontrolled experiment of geoengineering by altering the Earth's albedo (Boley and
2185 Byers, 2021).

2186
2187 Additionally, fragmentation events like explosions, impacts and loss of mass (solid rocket
2188 motor dust, paint flakes and other ejecta), or deliberate destructions lead to a continued
2189 growth of the number of space objects parallel to the increasing overall mass (Lawrence et
2190 al., 2022). Fragmentation is a constant source of sub 100 μm particles in LEO, which are
2191 extremely numerous (see Table 2.5). It is currently unclear how far down the size range of
2192 these sources extend. It is also unclear whether these particles undergo further processing
2193 and transport to contribute as a heavy ion source before particles are lost from the system
2194 due to atmospheric drag or solar radiation. Space missions measuring dust in this size
2195 range in LEO, like dust instruments on board *Destiny+* asteroid mission (Krueger et al.
2196 2019), are vital in studying these questions.

2197
2198 Finally, the increase in rocket launches might have an effect on ion transport from
2199 mesospheric heights due to gravity and acoustic waves generated by rocket exhaust (e.g
2200 Noble 1990; Mabie et al. 2016), although its effectiveness is unclear.

2201

2202

2203 5. Summary and Future observation

2204

2205 5.1 Summary of unanswered science questions

2206

2207 For high charge-state heavy ions that obviously originate from the solar wind (including
2208 solar energetic particles), there are some statistics for each large-scale region as shown in
2209 Figure 1.1, but even the basic entry route, the entry mechanism (similarity and difference
2210 from the alpha particle), and energization level from the original source are still
2211 unanswered. This is mainly due to the severe limitation of the existing instruments for such
2212 measurements. So far Geotail/STICS provided the best dataset for high charge-state
2213 heavy ions in the magnetosphere and its surroundings, but the instrument capability is
2214 limited to high energy (> 100 keV) and to very low time resolution for heavy ions.
2215 Therefore, we could not even separate between the direct effect of the solar wind and
2216 magnetospheric activity that influence the configuration of the magnetospheric boundary.

2217

2218 The same problem applies to the low charge-state metallic and heavy molecular ions,
2219 which have several candidates as the source. We here stress that their extremely low flux
2220 (no single source is hiding the other source like H^+ or O^+) in turn gives unique information
2221 of the source and the supply route as the tracer. In addition, even the sources are not
2222 determined: the Moon and/or the Earth's upper atmosphere, and for the upper atmosphere
2223 a fraction of the ions coming from the nightside sub-auroral region compared to the
2224 dayside polar region. The only route so far established is dayside high-latitude (through
2225 the plasma lobe) for heavy molecular ion that is the same route as one of the routes for O^+
2226 circulation and hence is expected to take the similar route and energization until they
2227 reach the inner magnetosphere (Yamauchi, 2019, and references therein). The other
2228 routes are difficult to identify, and it is even more difficult to evaluate the effect of the
2229 external conditions (e.g., solar wind, magnetospheric activity, Moon phase) under which
2230 ions from these sources appear.

2231

2232 Nevertheless, by re-examining the existing data from instruments that were not designated
2233 for separating molecular ions (e.g., Cluster/CODIF), and combining with new data (e.g.,
2234 from e-POP), we could identify new direct supply routes of the molecular ions (auroral
2235 region) to the inner magnetosphere, which is independent from the known high-latitude
2236 route.

2237

2238 Re-examination of existing data (e.g., Geotail/STIC and ACE) and combining other
2239 datasets (e.g., Kaguya and Chang'e) is also important in evaluating the Moon source as
2240 discussed in Sect. 4.1: We cannot rule out the possibility of a substantial contribution from
2241 the Moon during CIR/CME time periods. Thus, we still do not know the relative importance
2242 among the Moon, mesosphere, or the solar wind as the source of the low-charge state
2243 metallic ions. In this respect, further examination of Kaguya (mission terminated in 2009) is
2244 important as well as continued analyses of on-going e-POP and Arase, all of which are
2245 capable of detecting molecular ions and/or metallic ions.

2246

2247 For the other possible clues to investigate the upflow of the metallic ions, new models like
2248 WACCM-X and SAMI show that metallic ions might be transported regularly to altitudes
2249 above 500 km (above the exobase) in the particular locations (high-latitude). These models
2250 should ideally be combined with ionosphere-magnetosphere models and with possible
2251 observations in future (see Sect. 5.3 below). Such model-model and model-observation
2252 comparisons will further determine the probability that these heavy metallic ions can be
2253 lifted into the magnetosphere. Finally, contamination by space debris/waste emerges as a
2254 new open question, requiring new works in the future.

2255

2256

2257 **5.2 Desired specification for observation and model**

2258 Since our observational knowledge on the heavy molecular and metallic ions ($m > 20$) in
2259 the magnetospheric is very poor, we first need to define what measurements are highest
2260 priority for these ions. The lack of dedicated missions in the past is partly because it has
2261 been difficult to design ion instruments that can separate heavy species with sufficient
2262 geometric factor. To design the optimum set of feasible instrumentation, we need to know
2263 the limitation of the ion instruments on board past and current spacecraft. The limitations
2264 are:

2265

- 2266 1. In energy coverage: e.g., the IRM ion mass spectrometer onboard e-POP was limited to
2267 ions up to 90 eV/e (Yau and Howarth, 2016), and the STICS ion mass spectrometer
2268 onboard Geotail had an energy threshold of ~ 10 keV (Williams et al., 1994).
- 2269 2. Ambiguity of ion trajectory with finite entrance cross-section and small deviation of
2270 electric and magnetic field inside the instrument reduce the mass resolution: e.g., the

2271 CODIF ion mass spectrometer onboard Cluster had a mass resolution of $m/\Delta m \approx 5-7$
 2272 (Rème et al., 2001), and HPCA onboard MMS had $m/\Delta m \approx 4$ (Young et al. 2016).

2273 3. Size of the instrument and upper limit of the field strength inside the instruments limit
 2274 the mass range.

2275 4. Fragmentation of the molecular ions when going through the thin carbon foil limits the
 2276 molecular ion measurements for foil-type TOF instruments (e.g., Heredia-Avalos and
 2277 Garcia-Molina, 2000).

2278 5. Small geometric factor, which requires long integration time to accumulate adequate
 2279 counting statistics for minor species (e.g., Haaland et al., 2020). Currently, obtaining
 2280 angular (pitch angle) information within a short sampling time and at high mass
 2281 resolution is not easy.

2282 6. The instrument sensitivity degrades over the years due to the detector gain fatigue
 2283 mechanism (e.g., Kistler et al., 2013).

2284 7. Gaussian (as opposed to exponential) line shape of instrument mass response results
 2285 in non-negligible contributions to the measured minor ion counts from nearby major ion
 2286 species (cf. Figure 4.7).

2287
 2288 As a result, even the separation of the molecular ion measurements (separating $m/q \approx 30$
 2289 from $m/q = 16$ without charge state information) has been difficult for medium energy (50
 2290 eV – 50 keV), for which Arase was the first magnetospheric mission that can separate the
 2291 molecular ion group at the medium-energy. The instrument onboard Wind aimed the
 2292 resolution in design, but the achieved resolution was not sufficient due to technical issues.
 2293 Therefore, unlike traditional ion mass analyzers for the four major magnetospheric species
 2294 (the H^+ , He^{++} , He^+ , and O^+ group), we cannot require sufficient resolution for all directions
 2295 (temporal, energy, angular) when measuring the metallic and heavy molecular ions.

2296
 2297 With these limitations in mind, we have to define the direction of instrumentation and
 2298 measurement improvements to advance our knowledge.

2299 1. Have a mass range up to > 60 amu, to include Fe ions and heavy ionospheric molecular
 2300 ions (see Figure 1.1).

2301 2. Cover most important energy ranges that are different in different magnetospheric
 2302 regions: from cold ions (eventually few eV by ram flow) up to < 100 eV corresponding to
 2303 the upwelling ions at low altitudes below the main energization region (Yau et al., 2021),
 2304 from a few eV up to a few keV corresponding to the outflowing heavy ions at high

altitudes above the main energization region or the cusp (Kistler et al., 2010a), up to few 10's of keV corresponding to energetic heavy ions streaming down tail (Christon et al., 1994; Seki et al., 1998), and to a higher energy range for ions in the ring current, where the returning ions experience adiabatic acceleration (Ejiri, 1978). The wide energy range is also needed in the magnetospheric boundary region where pickup cold heavy ions and foreshock heavy ions can gain energy (Grünwaldt et al., 1997, Stasiewicz et al., 2013).

3. Obtain modest angular resolution with respect to the magnetic field while providing a mass resolution $m/\Delta m \geq 15$ over a wide energy range. This allows separating minor molecular ions ($m = 25 - 35$) from CNO-group atomic ions ($m < 20$), and minor heavier metallic ions ($m > 40$) from molecular ions. Such a mass resolution can be achieved through an isochronous TOF ion mass spectrometer, such as e.g. the MSA instrument onboard BepiColombo MMO (Delcourt et al., 2016) and JDC instrument onboard JUICE (Wittmann, 2022), or through a grazing incidence MCP time-of-flight ion mass spectrometer (Devoto et al., 2008). Alternatively, for ions up to keV, a magnetic ion mass spectrometer such as the IMA instrument onboard Mars Express (Barabash et al., 2006) would achieve this requirement by tuning the mass range and size (Nicolaou et al., 2017). The advantage of this design is that the ions do not touch the instrument elements other than the final detector (no fragmentation nor loss of energy).

4. Employ a high geometric factor design, or a new filtering/starting TOF mechanism (against the fragmentation of the molecular ions at the starting foil/surface of TOF) to allow for the separation of molecular ions within 30 minutes for keV range and 10 minutes for cold ions.

5. Employ a high mass resolution design of $m/\Delta m \geq 60$, to separate Al^+ ($m = 27$) from Si^+ ($m=28$) or N_2^+ ($m = 28$) at least above the ionosphere and in the ring current. The CELIAS/MTOF instrument on the SOHO spacecraft provided such a mass resolution for solar wind ions, i.e., for 0.5 - 30 keV/nuc ions (Hovestadt et al. 1995). There are designs and prototyping for such instruments that allow even for $m/\Delta m > 100$ (Wurz et al., 1998). Rosetta was equipped with a design for cold (<10 eV) ions (Balsiger et al. 2007). The technology is also available for hot (0.05-10 keV) ions on Kaguya/IMA instrument (Saito et al., 2010), i.e., $m/\Delta m > 15$, and improved to $m/\Delta m > 40$ for Mio (launched 2018), and $m/\Delta m > 100$ for MMX (to be launched in 2026).

6. Time resolution must be sufficient to separate different possible supply routes (e.g., the direct supply from the ionosphere and detoured supply through the tail) and to complete

2339 the traversal over each region. Here, the mass resolution and time resolution are
2340 related, and higher energy ranges generally require longer integration time due to the
2341 much lower ion flux in the Earth's magnetosphere. If we require as low as $m/\Delta m > 15$,
2342 instruments that fulfil this requirement (tens of minutes to hours to move from one
2343 region to another for high-altitude mission) are available almost all energy ranges.

2344
2345 The monitoring of the heavy ions is needed both in the magnetospheric region and in the
2346 possible source region. While the solar wind is being monitored at L1 for space weather
2347 monitoring purposes, new monitoring spacecraft or base is need for the Moon and the
2348 ionosphere/mesosphere. Here, the ionosphere includes both lower altitudes and above the
2349 exobase. To correlate any "event" in the source region and magnetospheric observation,
2350 time resolution must be comparable or less than such an "event", which means a time
2351 resolution < 10 minutes (e.g., substorms) for ionospheric origin low energy ions and hours
2352 for Moon origin ions or magnetospheric high-energy ions.

2353
2354 On the other hand, the duration of heavy ion events in the magnetosphere can be as long
2355 as a few days, i.e., the time scale of magnetic storms. Also, CME and CIR last several
2356 hours to a day. The question is then whether the heavy ions are supplied intermittently or
2357 continuously, or simply at one time with a slow and long decay. To distinguish between
2358 them, we need continuous monitoring (i.e., to return to the same region) for many days in
2359 raw. These time-scale requirements limit the types of acceptable spacecraft orbits.

2360 2361 **5.3 Desired missions and observations**

2362
2363 For in-situ observations, the best is to have dedicated mission, even as a secondary
2364 objective of any mission. Alternatively, placing a set of instruments on non-science
2365 missions such as Earth Observation satellites and Space Safety missions (including
2366 geostationary one) or the transfer spacecraft to/from the coming Lunar Gateway. The
2367 current plan for the Lunar Gateway is to be equipped with HERMES (Heliophysics
2368 Environmental and Radiation Measurement Experiment Suite) instrument package,
2369 comprising also the SPAN-Ai Ion Mass Spectrometer (2 eV - 40 keV ions, although $m/\Delta m$
2370 about 10 at moment).

2371

2372 In all cases, the key instruments are (1) an ion mass spectrometer of $m/\Delta m > 60$ for wide
2373 mass range of $m/q > 60$, and (2) lower mass resolution (yet $m/\Delta m > 15$) but sufficient
2374 angular resolution of at least 22.5° (6° is ideal because, for example, this is needed to
2375 detect the position where the ions are generated around the Moon), while keeping mass
2376 range up to $m/q > 60$. (3) In addition, adding total ion flux without mass resolution but with
2377 very good $\Delta E/E$ is good complement for absolute accuracy in velocity and particularly the
2378 density. The energy range of the instruments are divided into (a) cold, (b) hot up to few
2379 tens keV, and energetic. For the hot ions, it would be wise to divide into two different
2380 energy ranges to have better energy and mass resolution. In this case, it is also wise to
2381 use different detection methods for such divided energies each. In addition to the ion
2382 instruments, technology of the (4) optical limb observation from satellites has also
2383 advanced, e.g. sounding of Mg^+ . If limb observation becomes possible for the other
2384 metallic species, this would be a strong satellite tool in future.

2385

2386 Any of these key instruments are useful to be placed on the other missions mentioned
2387 above (as a package). As for the dedicated mission, it would be good to have a multi-
2388 spacecraft mission with the spacecraft having different orbits, such as low altitude (400 -
2389 4000 km), mid-altitude (2000 - 30000 km), and high-altitude (4 - 10 Re), as well as the
2390 ionospheric source (200 - 500 km). Monitoring of the possible source population is also
2391 important. For the solar wind source, L1 monitoring spacecraft has been and will provide
2392 key information, whereas for the Moon source, the coming Lunar Gateway would be a
2393 good platform to place the key instrument like Kaguya/IMA.

2394

2395 If allowed only one spacecraft, then that mission should aim for all escaping ions, and
2396 have a highly elliptic because most of the Earth observation satellites are in nearly circular
2397 orbit, such as the ESCAPE (European SpaceCraft for the study of Atmospheric Particle
2398 Escape) mission, which was proposed to ESA in response to the M5 call (Dandouras et
2399 al., 2018). The proposed ESCAPE mission spacecraft was designed as a slowly spinning
2400 spacecraft on a high inclination $500 \text{ km} \times 33\,000 \text{ km}$ orbit, i.e., with a perigee at the
2401 terrestrial exobase, and was equipped with instrumentation responding to the above
2402 criteria. With many ion mass instruments with an optical monitor of the exospheric heavy
2403 atoms, it was designed to separate even nitrogen and oxygen. With modern technology,
2404 several dips into much lower altitude to cover the middle thermosphere and ionosphere

2405 could be considered, similar to what MAVEN satellite did for Mars with the Neutral Gas
2406 and Ion Mass Spectrometer (NGIMS) instrument onboard.

2407

2408 The monitoring of the source region is also essential. Since the Moon monitor is
2409 mentioned in previous subsection, we here describe the Earth part (ground-based
2410 observations) including the ultimate source of meteorites. Here, the important outputs are
2411 how much of the source ions may reach the exobase and topside ionosphere. This
2412 requires, in addition to monitoring the geomagnetic activity that is already available, (1)
2413 instruments and models that provide upward transport of heavy ions in the mesosphere,
2414 ionosphere, and thermosphere with high spatial resolution, and (2) monitoring of possible
2415 source population (although it is only "possible" source with current understanding) of the
2416 amount of metallic ions and atoms in the lower part of the ionosphere. Candidate for (1) is
2417 modern incoherent scatter radar such as EISCAT_3D, and candidate for (2) is lidars.

2418

2419 For lidars, we need to keep the observation site, and even expand to more sites (ideally
2420 two latitudes at $\sim 80^\circ$ and $\sim 60^\circ$ in both hemispheres corresponding to two major outflow
2421 latitudes that e-POP revealed, and three longitudinal locations to separate temporal and
2422 diurnal variations) as well as target species. We particularly need to monitor Ca^+ layer
2423 (ionized form) and Fe layer (originated from mainly meteoroids but space debris/waste
2424 might become detectable first in amount), while searching for AIO (the Al species
2425 observable by lidar) could be useful for evaluating the contribution from the space
2426 debris/waste, although its current AIO density predicted by an atmospheric model is very
2427 low (Plane et al., 2021)

2428

2429 Since it is unclear how much transient enhancements of the metal layer influence the total
2430 upward transport (such as by meteor showers and large re-entry events of space
2431 debris/waste) it is desirable to gain a more complete picture of the current input of mass to
2432 the atmosphere. This also includes the observation of the ablation process, by which the
2433 material is distributed in the atmosphere. These observations should be performed using
2434 many different detection methods (e.g. dust detectors, lidars, radars, optical systems) and
2435 missions.

2436

2437 The observation of the ablation process is particularly important for evaluating the ablation
2438 of space debris/waste because the modelling and indirect observations suggest that minor

2439 species such as Al, Cu, Ge, Li and Pb coming from space debris/waste might already
2440 constitute a significant contamination to the naturally occurring particles from meteoroids.
2441 There are currently no data on very small-scale debris statistics and evolution. For
2442 evaluating this effect, a multi-probe experimental flight through a possible ion cloud (after
2443 re-entry of the first probe that imitates the space debris/waste) would be useful. Such an
2444 experiment would gather information on the ablated amount and altitude distribution, as
2445 well as the effectiveness of vertical transport of the generated ion cloud. For such an
2446 experiment, remote observation from the LEO satellite should also be added by aligning
2447 the re-entry orbit to the ideal LEO satellite for monitoring. As a similar attempt, Deadalus
2448 proposal (Sarris et al., 2020) is can make such attempt although the target is ionosphere
2449 but not mesosphere.

2452 **5.4 Modelling of upward transport of metallic and molecular ions in the ionosphere** 2453 **and mesosphere**

2454
2455 It would be ideal if one can assimilate the relatively rare observations (e.g., in Sect. 4) into
2456 thermospheric and ionospheric transport models. The molecular ion upflow at sub-auroral
2457 latitude and sudden expansion of the metal layer (see the example in Sect. 4.3.2 for Na)
2458 are examples that need to be reproduced by transport models.

2459
2460 For modelling (particularly for evaluating the ablation of space debris/waste), the source
2461 flux for the solar wind and the upper atmospheric molecular ion distribution as the ground
2462 state are well understood. Also, in the case of the mesospheric metal layers, the current
2463 understanding is quite mature, and we have a long history of measurements and models.
2464 As such, for the purpose of transport to the magnetosphere, the mass input to these metal
2465 layers by meteoroids is no longer required in modelling except for the increasing input from
2466 the ablation of the space debris/waste.

2467
2468 On the other hand, understanding the transport of heavy metals from the mesosphere to
2469 higher altitudes needs further understood because of lack of monitoring satellites for these
2470 heavy ions for past 40 years as summarized in Table 3.1. The last capable satellite was
2471 DE-2 in early 1980's and flight much higher altitude than exobase. Alongside such
2472 additional observations, model development is also needed to evaluate the transport of

2473 metallic ions to beyond the exobase. Currently WACCM-X reaches 500 km altitude but not
2474 further. One possible development could be to couple such atmospheric and ionospheric
2475 models with magnetospheric models of wave-particle interaction. This would extend the
2476 modelling regime into the magnetosphere and would yield predictions that could be
2477 compared with measurements.

2478

2479 Although the meteoric mass input to the atmosphere is roughly known and it is possible to
2480 estimate the contribution from the re-entry of human-made objects (e.g., satellites and
2481 space debris), the details concerning the ablation needs more modelling and observations,
2482 and total influx estimates and its composition need refining. These details are needed in
2483 order to estimate in what way and how much these influxes contribute to the mesospheric
2484 metal layer. This is particularly important for space debris/waste to prognose the future
2485 evolution of the metal layer by the increasing launch of rockets to the space. Almost no
2486 observational work has been done on examining the deposition altitude distribution and
2487 composition from deorbiting debris. Thus, comprehensive modelling of the ablation
2488 process of space debris/waste is needed. Due to the complex nature of the ablation and
2489 the subsequent chemical and physical processing of the injected material, modelling
2490 should be complemented and validated by more detailed and numerous observations of
2491 spacecraft re-entries, especially of spacecraft resembling typical orbital and mass
2492 characteristics of the already numerous low-Earth orbit communication satellites (LCS).
2493 Although there are estimates on the total influx of debris based on the mass of objects
2494 contained in regions that will deorbit, the deposition altitudes are key to understanding the
2495 impact on our atmosphere. In some areas it might be time-critical to intensify the research
2496 before the anthropogenic influence due to the increase in re-entry of space debris/waste
2497 reaches a level that causes significant perturbations to natural environment in the
2498 atmosphere and the space.

2499

2500

2501 **6. Conclusion**

2502

2503 Very minor ions in the magnetosphere have unique information because of their low flux
2504 (no single source is hiding the other source like the case for H^+ or O^+), on both the source
2505 and the transport route. However, their transport is not well understood because of the

2506 lack of observations, which partly results from the instrument capabilities for the terrestrial
2507 missions in the past and present. While we need new observations with suitable
2508 instrumentation, the technology and design of which are already available and are used in
2509 current planetary missions, re-examining the exiting datasets, and combining different
2510 datasets can also give new insights on this subject. We have shown several examples for
2511 such re-examinations of data, e.g., the importance of considering the Moon and space
2512 debris/waste as the detectable sources for the metallic ions in the magnetosphere.
2513 Therefore, a related database of measurements taken by old instruments listed in section
2514 3 with designs more than 20-years old are still useful, along with more recent models. This
2515 in turn means that we need new observations and missions in the near future with
2516 available technology, as summarized in section 5.

2517

2518

2519

2520 **Acknowledgments**

2521

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