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IRON ATOMS AND METALLIC SPECIES IN THE EARTH'S UPPER ATMOSPHERE

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Abstract. Resonant lidar detection of iron atoms have been performed at the Observatoire de Haute Provence (France) during 4 nights in november 1986 and in april 1987. The average iron atoms abundance is 3.6×10^9 cm^{-2} in november, and 2.6×10^9 cm^{-2} in april. Iron atoms density profiles are compared to the atomic sodium ones, obtained simultaneously by lidar and indicating an average sodium abundance of 4×10^9 cm^{-2}. The relative abundance of the sum of the atomic and ionic forms is compared for several metallic species with their abundance ratios in the incoming meteorites. Finally, similar comparisons using lidar and mass spectrometer data are made for the ratio of the atomic and the ionic forms in the atmosphere.

Introduction

Metallic species present in the earth upper atmosphere at the mesopause level have been studied for many years, the most likely hypothesis concerning their origin being related to the vaporization of the meteorites entering the earth's atmosphere. Among these metallic species, only the alkali ones (Na, K, Li) have been measured on a routine basis, first by photometry (Donahue and Blamont, 1961 ; Gadsden, 1969; Gault and Rundle, 1969), and, since 1969, by laser soundings (Gibson and Sandford, 1971 ; Megie and Blamont, 1977 ; Simonich et al., 1979 ; Megie et al., 1978 ; Jegou et al., 1980). The results of these experiments have been reviewed by Megie (1988). More recently, lidar observations of atomic calcium have been obtained over a period of 5 years at the Observatoire de Haute Provence (O.H.P.), from 1982 to 1987 (Granier et al., 1988). All these observations have demonstrated the existence of the metals in the atomic form in the 80-110 km altitude region.

Rocket-borne mass spectrometers measurements have also shown the presence of several metallic ions - Mg^+, Fe^+, Si^+, Na^+, Ca^+ - in the same altitude range, but, up to now, no systematic data concerning these ionic species were available. Lidar observations of ionic calcium have been obtained at the O.H.P. over a 2 years period in 1983 and 1984, for the summer and autumn seasons (Granier et al., 1988).

Many theoretical studies have focused on the behavior of these species and their compounds at various temporal scales ; these studies (Brown, 1973 ; Richter and Sechrist, 1979 ; Jegou et al., 1985) show that, above 85 km, the preponderant forms are the atoms and the atomic ions. Validations of such models could be achieved by comparing, for various metals, the experimental and theoretical results concerning both the absolute abundance of each compound and the abundances ratio between the atomic and ionic form of each species.

Up to now, such comparisons are limited to the alkali and calcium compounds. But these metallic species, Na, K and Ca are only minor components of the meteoritic source (about 1% in weight), the main ones being Mg, Si and Fe. Concerning the atomic form of the last three, only few detections of the iron atoms have been done by twilight photometry, first by Broadfoot and Johanson (1976), who measured an atomic iron total content of about 7.10^8 cm^{-2} above 90 km. Using the same technique, Tepley et al. (1981) and Mathews (1981) have measured a maximum atomic iron content of 10^9 cm^{-2} below 90 km. The analysis of these data has been theoretically discussed by Swider (1984), who considers these abundances to be strongly underestimated. The atomic iron content remains today uncertain by at least a factor of ten.

New measurements of the iron atoms density profiles using a ground-based lidar system located at the Observatoire de Haute-Provence (O.H.P., 44°N, 6°E) will be presented here. It can be noted that, among the three main metallic components of the meteoritic source, only the atomic form of iron can be detected by lidar from the ground, the resonant wavelengths of the two other being strongly absorbed in the ultraviolet range, below 300 nm.
The resonance line a^5D-z^5F has been chosen for the measurements, which corresponds to a wavelength of 372 nm. The characteristics of this line - Doppler broadened linewidth, oscillator strength and peak cross-section - are given in table 1 (Fuhr et al., 1981). In order to compare these values with those of the species previously detected by lidar, the spectral features of the atomic Na and Ca resonance lines are also given in this table : the value of the atomic iron cross-section is about 20 times below the Na and Ca ones.

Experimental set-up

The lidar system used for the experiments, presented on figure 1, is similar to the one previously used for the ionic calcium detection (Granier et al., 1985). The emission at 372 nm is obtained by a frequency mixing process. The infra-red emission of the Nd:Yag laser remaining after frequency doubling and the tunable emission of a dye laser around 572 nm are mixed in a KDP crystal, which provides the atomic iron resonance wavelength.

The characteristics of the experimental systems (emitter and receiver) are summarized in table 2. The emitted energy is about 15 mJ at a 10 Hz repetition rate. The dye solution used is Rhodamine 6G in methanol, which allows to maintain this energy over a 60 hours period. The determination of the wavelength is made by using a 0.5 pm resolution spectrometer (Sopra, model F1500), the reference being given by a hollow-cathod lamp. A more precise tuning of the wavelength can be achieved by scanning the atmospheric resonance line from the ground (Granier et al., 1985).

Considering the total efficiency of the system, the detection thresholds have been calculated, assuming they correspond to a signal to noise ratio of 5, and for a typical 4 hour integration time during nighttime and daytime periods. They are given in table 3, with the corresponding values for the

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Table 1: Characteristics of the resonance lines of Fe, Na and Ca atoms.

	Fe	Na	Ca
Transition	$a^5D - z^5F$	$^2S_{1/2} - ^2P_{3/2}$	$^1S_o - ^1P_1$
Wavelength (nm)	371.9930	588.9953	422.6728
Doppler width at 190 K (pm)	0.49 pm	1.21 pm	0.66 pm
Oscillator strength	0.0413	0.655	1.75
Peak cross-section (cm^{-2})	$9.47\ 10^{-13}$	$1.52\ 10^{-11}$	$3.85\ 10^{-11}$

sodium atoms. The lidar measurements are, up to now, limited to the nighttime period. The detection limit correspond to a iron atoms total content of 10^8 cm^{-2}, about 10^2 times above the corresponding sodium ones : such a limit allows a precise determination of the iron atoms total content, as the previous photometric measurements have shown values above 7.10^8 cm^{-2}.

For the atomic iron measurements, as for the other metallic species lidar measurements performed before, the main errors reside in the evaluation of the statistical fluctuations of the signal and of the effective cross-section of the resonant process ; the last one results mainly from the errors in the fine determination of the emitted wavelength, and of the spectral width of the laser line (Granier et al., 1985). Taking into consideration all the parameters, the total errors on the density profiles and abundances can be evaluated to ± 20% (Mégie, 1988).

Table 2 : Characteristics of the lidar system used for the iron atoms detection

Emission		**Reception**	
Wavelength	372 nm	Telescope diameter	0.8 m
Output energy	15 mJ	Filter bandwidth	3 nm
Laser linewidth	1.8 pm	Filter transmission	12%
Beam divergence	10^{-3} rad	Sampling gate	4μs
Repetition rate	10 Hz		

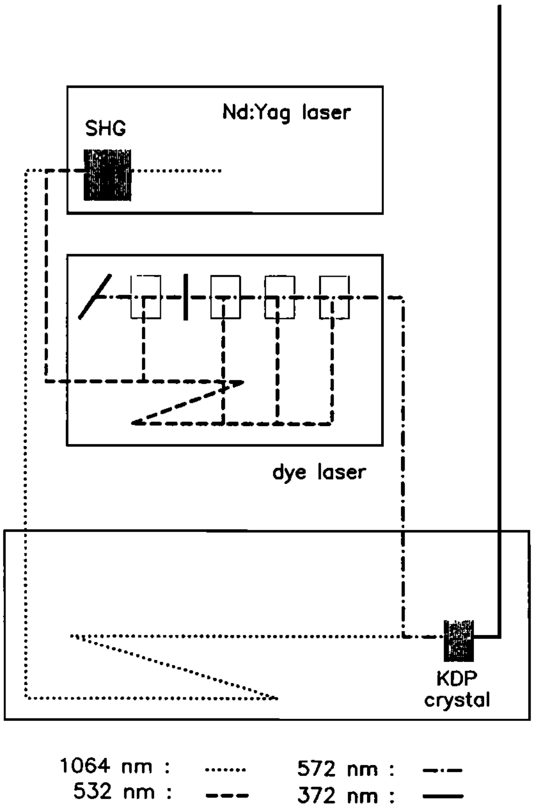

Fig. 1 : Schematic configuration of the emission system.

Experimental data

The first measurements of the iron atom density profiles have been performed on November 18, 1986. Measurements of this species are now available over 4 nights, 3 in November 1986 (18, 20 and 22), and one on April 27, 1987. During the 3 nights in November, the sodium atoms have also been detected, alternately with the iron ones. During these nights, iron atoms density profiles have been obtained with a time resolution of about 20 minutes.

The average atomic iron density profiles for each of these nights are plotted on figure 2, together with the sodium ones, when available. The average iron atoms content for the 3 nights of november 1986 is $3.6\ 10^9$ cm^{-2}, the one obtained in april being $2.6\ 10^9$ cm^{-2}. Such values are larger than the abundances measured by Broadfoot and Johanson (1976) and by Tepley (1981), but remain largely lower than the theoretical ones estimated by Swider (1984). The data obtained in November on sodium atoms show very similar abundances of the two elements: the mean sodium content, 4.10^9 cm^{-2}, is only 10% larger than the iron one.

Furthermore, when considering the altitude characteristics of the layers, as the layer width and the altitude of the maximum concentration, one can observe similar values for the iron atoms layer and those previously observed for the alkali and calcium atoms (Granier et al., 1988).

Discussion

Some experimental and theoretical studies (Gadsden, 1969 ; Zbinden et al., 1975 ; Swider, 1984) have tried to assess the meteoritic origin of the metallic species, by comparing the ratios of the

Table 3 : Detection thresholds for a 4 hour integration time

		Concentration	Total abundance
Night			
	Fe	110 cm^{-3}	$1.1\ 10^8$ cm^{-2}
	Na	0.8 cm^{-3}	$8.0\ 10^5$ cm^{-2}
Day			
	Fe	2100 cm^{-3}	$2.1\ 10^9$ cm^{-2}
	Na	15 cm^{-3}	$1.5\ 10^7$ cm^{-2}

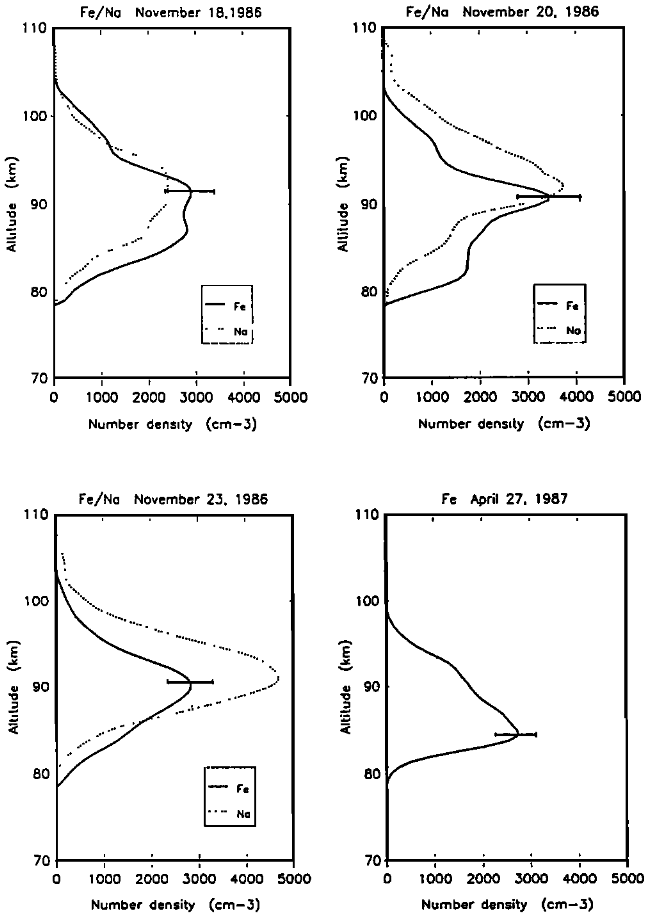

Fig. 2 . Average atomic Fe and Na density profiles for each night of Fe measurements.

different metallic compounds in the atmosphere and in the meteorites.

Up to now, due to the lack of data, these comparisons have been limited on one hand, to atomic alkali metals and on the other hand to the metallic ions. However, as seen previously, the sum of the atomic and ionic species abundances has to be considered for a better estimation of the total metal abundances in the 80-110 km altitude zone.

The mean available total contents of the metallic atoms and ions are given in table 4, for Na, K, Ca, Mg, Fe and Si compounds. The data concerning the Na, K, Ca, Ca$^+$ and Fe contents are average values obtained by lidar at the Observatoire de Haute-Provence. The data concerning the other metallic ions are average values deduced from two rocket flights performed at mid-latitude by Zbinden et al. (1975) and by Kopp and Hermann (1984). Although the first measurements were made at the maximum of the Geminid meteor shower, these values are representative within a factor of 2 to 5 of ion concentrations observed in the mid-latitude regions.

The relative abundance (Na=1) of the sum of the two forms, atom and ion, are compared in table 4 to their relative abundance in the meteorites, assumed to be mainly ordinary chondrites : the two alkali metals, Na and K, present nearly similar abundances in the atmosphere and in the meteorites. However, the normalized total iron content is about 8 times lower in the atmosphere than in the meteorites, while the normalized total calcium content is more than 70 times lower. For the two other major metallic compounds of the meteorites, no comparison can be done, due to the lack of data concerning the atomic form.

<u>Table 4</u> : Relative abundances of the metallic atoms and ions, and comparison with the ordinary chondrite ones.

Species	[M] (cm^{-2})	[M$^+$] (cm^{-2})	[M]+[M$^+$]/ [Na]+[Na$^+$]	ordinary chondrites
Na	3. 10^9	2 - 3 10^8	1	1
K	2. 10^8	5.10^6-4.10^7	1.5-8 10^{-2}	9. 10^{-2}
Ca	2.7 10^7	1 - 2 10^7	1.1-1.5 10^{-2}	1
Mg	?	1 - 2 10^9	> 0.3	22
Fe	3.6 10^9	2 - 3 10^9	1.2 - 1.8	17
Si	?	3 - 5 10^8	> 0.2	23

One can furthermore notice a large difference in the ratio between the atomic and ionic total abundances, which is in the range 15-40 for the alkali species, and between 1 and 4 for the calcium and iron ones.

Conclusion

The first lidar measurements of the atomic iron density profiles and total abundances performed on four nights - november 1986, april 1987 - have given new information on the atmospheric chemical equilibrium of one of the major components of the source of the atmospheric metallic species.

Comparisons of these data with ionic iron mass-spectrometer measurements, and with Na-Ca-Ca$^+$ lidar measurements performed on the same site show that on one hand the alkali, and on the other hand, the other metallic species, as calcium and iron, have very different behaviors, concerning both the total abundance with reference to the meteoritic source and the abundance ratio between the atomic and ionic forms.

One consequence of these new measurements seem to be the need for a clear difference between the alkali chemistry and the calcium (alkaline-earth) and iron (transition metal) ones : the ion - neutral coupling might be different, as well as the source ratios.

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