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# Measurement and modelling of gaseous elemental iodine (I<sub>2</sub>) dry deposition velocity on grass in the environment

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## Abstract

Assessing the impact of radioactive iodine on humans subsequent to a nuclear accident requires a better understanding of its behaviour in the environment. An original approach aimed at developing a model constrained by data collected during experimental campaigns has been developed. These experimental campaigns, named MIOSEC 2 and MIOSEC 3 respectively, were conducted in the middle of grassland. They are based on emissions of gaseous elemental iodine (I<sub>2</sub>) into the atmosphere above the grassland to determine the dry deposition velocities of iodine on the grass and to model these velocities as a function of the environmental conditions, particularly wind friction velocity, sensible heat flux, and stomatal resistance. The measured dry deposition velocities were between 0.02 and 0.49 cm.s<sup>-1</sup> during MIOSEC 2, varying by more than one order of magnitude, and between 0.48 and 1.25 cm.s<sup>-1</sup> during MIOSEC 3. The dry deposition model for iodine developed as a result of these experiments relies on the micrometeorological characteristics of the atmospheric surface layer, the pertinent physical and chemical properties of the iodine and the surface properties of the grass; all these parameters were measured at the time of the experiments. Given the experimental conditions, the modelled dry deposition velocities varied between 0.11 and 0.51 cm.s<sup>-1</sup> during MIOSEC 2 and between 0.31 and 1.6 cm.s<sup>-1</sup> during MIOSEC 3. The dry deposition model for iodine indicates that the variations in deposition velocity are induced by the mechanical turbulence, since there is significant correlation between the dry deposition velocities of iodine and the wind friction velocities on grass. The model also shows that the higher deposition velocity values during MIOSEC 3 are due to the fact

that the stomata were more open during the experiments. There is also significant correlation between the experimental results and modelled values both for MIOSEC 2 ( $R^2 = 0.61$ ) and for MIOSEC 3 ( $R^2 = 0.71$ ).

*Keywords:* Elemental iodine, environment, emission, grass, dry deposition velocity.

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## 1. Introduction

Iodine is a trace element in the halogen family. It is essential for thyroid function in animals. Iodine has 37 isotopes with mass numbers ranging from 108 to 144. Iodine-127 is the only stable isotope of iodine. All the other isotopes of iodine are radioactive. Thirteen are fission products, including iodine-129 which has the longest half-life (15.7 million years) and iodine-131 which has a half-life of 8.02 days. Most iodine is found in the marine environment (>70%), in dissolved forms at concentrations of between 45 and 60  $\mu\text{g.L}^{-1}$  (Fuge, 1996; Wong, 1991). It can also be found on land at concentrations influenced, among other parameters, by proximity to oceans, by the organic composition of the environment, by ground topography and by the bioavailability of molecules containing iodine. The mechanisms of iodine transfer into the atmosphere from marine and terrestrial environments are not known, but the most likely is volatilization. In acid condition, the iodine present in seawater in ionic form is liable to volatilize as  $\text{I}_2$ , whereas in neutral or alkaline condition, volatilization is unlikely (Fuge, 1996; Hou et al., 2009). In addition, iodine can volatilize in the form of organic compounds, e.g.  $\text{ICH}_3$ , due to the oxidation of residues of plants such as seaweed (Fuge, 1996; Whitehead, 1984). The iodine ( $^{127}\text{I}$ ) concentrations measured in the atmosphere are between 10 and 100  $\text{ng.m}^{-3}$ , taking account of all physico-chemical forms (Truesdale et al., 2012). Other sources of atmospheric iodine ( $^{129}\text{I}$  and/or  $^{131}\text{I}$ ) are spontaneous uranium ( $^{238}\text{U}$ ) fission, nuclear testing, releases from spent fuel reprocessing plants and emissions from nuclear power plants in normal or accident operating conditions.

During a nuclear accident, radioactive iodine ( $^{131}\text{I}$ ) is released into the environment (Parache et al., 2011; Zhou, 1995). In the atmosphere, radioactive iodine diffuses in the same way as any other gas (Chamberlain and Chadwick, 1966). With oxidation states ranging from -1 to +5, the overall biogeochemical cycle of iodine is quite complex and involves various processes (Carpenter, 2003; Saiz-Lopez et al., 2012). Its high reactivity means that iodine, including radioactive iodine, changes rapidly in the environment through photolysis and reacts with other atmospheric compounds including ozone and nitrogen compounds (Carpenter, 2003; Hou et al., 2009; Noguchi and Murata, 1988). Because of this, radioactive iodine is present in the atmosphere in several physico-chemical forms: particulate (associated with atmospheric aerosol particles) and gaseous. The gaseous forms are inorganic:  $\text{I}_2$ ,  $\text{HI}$ ,  $\text{HIO}$ , and organic:  $\text{CHI}_3$ ,  $\text{CH}_2\text{I}_2$ ,  $\text{CH}_3\text{CH}_2\text{CH}_2\text{I}$ . Radioactive iodine concentrations can vary not only according to distance from the discharge stack (Thakur et al., 2013), but also according to season and climate when the radioactive half-life allows (Hou et al., 2009).

Subsequent to an accident, particular attention should be paid to the deposition mechanism of the iodine, which can be found in cows' milk via grazing, then in humans by milk drinking or by direct ingestion of fresh vegetables, where it can cause thyroid cancer. Deposition can occur in dry weather (dry deposition) or wet weather (wet deposition), and is quantified by velocity. Numerous studies have shown that the dry deposition velocities of iodine are highly dependent on the one hand on the state of iodine in the atmosphere: gaseous or particulate including in this last case the particle size, and on the other on the chemical species encountered (Nielsen, 1981; Noguchi and Murata, 1988). On grass, the dry deposition

velocity of gaseous elemental iodine measured in the environment by Chamberlain and Chadwick (1966) was  $1.8 \text{ cm.s}^{-1}$ , while Karunakara et al. (2018) measured dry deposition velocities of between  $0.5 \times 10^{-3}$  and  $3.3 \times 10^{-3} \text{ cm.s}^{-1}$  in an environmental chamber. These values differ by several orders of magnitude, since no parameterisation was done for meteorological values or surface properties of the grass.

This study aims to quantify and model the dry deposition velocities of gaseous elemental iodine ( $\text{I}_2$ ) on grass based on experiments conducted in the environment. The experiments consisted in emitting gaseous elemental iodine into the atmosphere during a limited period of time in the absence of precipitation and quantifying the quantities of iodine deposited on grass test specimens placed downwind of the emission point. In case of rain during  $\text{I}_2$  emission,  $\text{I}_2$  can be absorbed by the rain drops, which will lead to modification of global transfer depending on the solubility of iodine and rain intensity (via drops size) and consequently, a modification of the deposition velocity. Because the average iodine concentrations in the atmosphere above the test specimens were known, dry deposition velocities were calculated and parameterised for wind friction velocity, sensible heat flux and stomatal resistance.

## 2. Methodology

### *A. Measurement of iodine dry deposition velocities on grass*

#### *1. Equations*

The iodine dry deposition velocities were calculated by dividing the iodine dry deposition fluxes by the atmospheric concentrations of iodine. For this purpose, the iodine dry deposition fluxes were obtained by dividing the iodine concentrations measured on the grass by the iodine emission time (Eq. (1)). The deposition velocities were then determined by dividing the mean iodine dry deposition fluxes on the 2 squares of grass for each of the experiments by the mean atmospheric iodine concentrations in the corresponding 2 bubbler flasks (Eq. (2)).

$$F = -C_d/t \quad (1)$$

Where  $F$  is the iodine dry deposition flux ( $\mu\text{g.m}^{-2}.\text{s}^{-1}$ );  $C_d$  is the iodine concentration deposited on the grass ( $\mu\text{g.m}^{-2}$ ) and  $t$  is the emission time (s).

$$V_d = -F/C_a(z) \quad (2)$$

Where  $V_d$  is the iodine dry deposition velocity ( $\text{m.s}^{-1}$ ) and  $C_a(z)$  the atmospheric iodine concentration ( $\mu\text{g.m}^{-3}$ ) at the height  $z$  at which the air is sampled.

To obtain the iodine concentrations in the air and on the grass, several experiments based on gaseous elemental iodine emissions were conducted on an experimental site in the middle of grassland.

#### *2. Description of the measurement site*

Two experimental campaigns, entitled MIOSEC 2 and MIOSEC 3, were conducted in the middle of grassland on an experimental site of the French National Institute for Agricultural Research (INRA) at Lusignan (France) ( $46^\circ 24', 02''\text{N}$ ;  $0^\circ 7'$ ,

04'E). MIOSEC 2 ran from 18 to 26 September 2018 and MIOSEC 3 from 4 to 7 June 2019. During MIOSEC 2, at any point approximately 500 m from the experiment, the grass was 0.10 m high and its mean roughness length ( $z_0$ ) was 0.01 m. During MIOSEC 3, at any point approximately 500 m from the experiment, the grass was 0.15 m high and its mean roughness length ( $z_0$ ) was 0.015 m (Table 1). The roughness lengths given are approximated on the basis of the grass height ( $z_0/z = 0.1$ ) (Pellerin et al., 2017). During the two measurement campaigns, the grass consisted mainly of *Lolium perenne* and *Dactylis*.

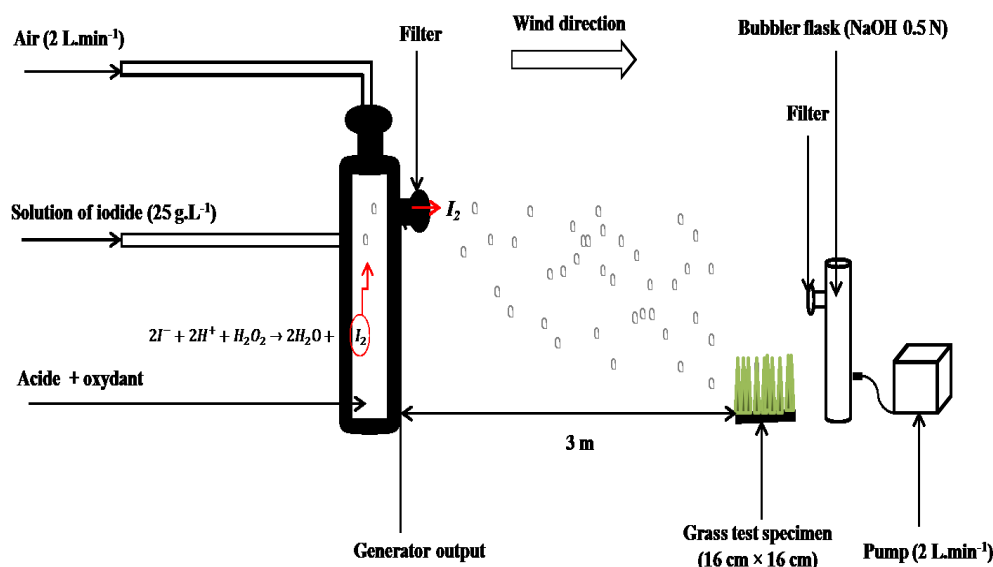
**Table 1:** Description of the measurement site.

Campaign (Date)	Type of cover (height)	Roughness length ( $z_0$ )	Surface homogeneity
MIOSEC 2 (18 to 26 September 2018)	Grassland (0.10 m)	0.01 m	> 500 m
MIOSEC 3 (4 to 7 June 2019)	Grassland (0.15 m)	0.015 m	> 500 m

### 3. Iodine emissions

The measurement campaigns use gaseous elemental iodine emissions produced using an iodine generator designed previously. To generate the iodine, 20 ml of a KI solution (25 g.L<sup>-1</sup>) are added drop by drop to a mixture of 100 ml of H<sub>2</sub>SO<sub>4</sub> (10%) and 100 ml of H<sub>2</sub>O<sub>2</sub> (35%) for 30 minutes. Each iodide (I<sup>-</sup>) droplet added to the mixture leads to the formation in solution of gaseous elemental iodine (I<sub>2</sub>). The gaseous form I<sub>2</sub> is entrained by a 2 L.min<sup>-1</sup> air flow running continuously for the 30-minute emission duration. A filter is placed at the iodine generator output to stop any iodine particles that might form inside the tube connected to the generator output (Fig. 1).

The homogeneity of iodine release from generator was validated during the generator design. Two flask each containing 100 ml of NaOH (0.5 N) are directly connected to the output of generator to trap iodine. In the event that the totality of iodine is not trapped in the first flask, the iodine excess is trapped in the second. Each 5 minutes, iodine generation is stopped, and the 2 flasks are replaced by 2 others. This operation is repeated 6 times, corresponding to the total duration of iodine emission (30 minutes). The measurement of the total iodine in the first flask validated the protocol for iodine trapping in 100 ml of NaOH (0.5 N). Iodine concentrations measured in the different flasks are the same. Consequently, iodine release from generator is homogeneous during the 30 minutes.



**Fig. 1.** Emission of gaseous elemental iodine, sampling from the atmosphere and grass.

#### 4. Collection and treatment of the samples (including blanks)

Two cellulose filters (0.45 µm) are placed between the emission point and the sampling points to collect any iodine particles that form during emission. Depending on the wind direction, 2 squares of grass (*Lolium perenne*) measuring 16 cm × 16 cm each are placed 3 m from the generator output to collect the deposited iodine. Each experiment being carried out during 30 minutes and “field loss half-life” for elemental iodine being determined (13 days) on grassland in the growing season (Whitehead, 1984), re-volatilization is therefore very low during 30 minutes and has not been determined.

Two flasks, each containing 100 mL of NaOH (0.5 N) are placed approximately 5 cm from the squares of grass to collect the atmospheric iodine, by means of sparging at a rate of 2 L.min<sup>-1</sup> using a pump connected to the flask output (Fig. 1). A filter is placed at the input of each bubbler flask to collect any iodine particles. 14 and 8 experiments were conducted during MIOSEC 2 and MIOSEC 3, respectively. After each experiment, the sample were collected and treated.

For each of the experiments, a grass blank and an atmospheric blank were made by placing at 50 m from emission point, a 16 cm x 16 cm square of grass and a bubbler flask containing 100 ml of NaOH (0.5 N), respectively.

The particulate iodine fraction was collected by placing each of the filters in 50 ml of NaOH (0.5 N). After vigorous homogenisation, the whole mixture was filtered.

To determine the quantities of iodine emitted into the atmosphere, the samples collected from the bubbler flasks were filtered. The atmospheric blank was treated in the same way as the samples from the bubbler flasks.

The quantities of iodine deposited were determined from the grass. The squares of grass were cut so as to collect the maximum mass. To extract iodine from grass samples, an original methodology was developed. Iodine was separated from grass using calcination at 400 °C (Table 2) followed by liquid-liquid extraction (Fig. 2). Firstly, each square of cut grass was placed in a flask containing 150ml of NaOH (5 N). A physico-chemical treatment was then applied to the samples. The

149 samples were placed in porcelain crucibles and calcined at 400°C to destroy organic matter. After cooling, each sample was  
 150 dissolved in 150 ml of ultrapure water (18.2 MΩ cm at 25°C) and then filtered. Iodine is recovered from the filtrate as  
 151 iodide form. H<sub>2</sub>SO<sub>4</sub> (30%) was added to adjust pH to 2 and H<sub>2</sub>O<sub>2</sub> (35%) to oxidize iodide to iodine. After agitation and  
 152 settle down until phase separation, iodine in solution is released and trapped by trimethylbenzene as I<sub>2</sub> form. The aqueous  
 153 phase was eliminated and the phase containing the trimethylbenzene was isolated and washed with ultrapure water until its  
 154 pH was neutral. Then 100 ml of NaOH (0.5 N) was added to extract the iodine in the form I<sup>-</sup> (back extraction). The grass  
 155 blank was treated in the same way as the grass samples (Fig. 2).

156

157 **Table 2:** Calcination cycle to separate the iodine (<sup>127</sup>I) from the grass.

Step	Temperature (°C)	Duration (min)
1	25-100	75
2	100-100	120
3	100-200	100
4	200-200	120
5	200-400	200
6	400-400	120
7	400-25	375

158 To determine the extraction efficiency, grass blank and 12 tests were performed. For each test, 5 ml of KI (1g.L<sup>-1</sup>)  
 159 were added to 20 g of fresh grass. After adding 150 ml of NaOH (5 N), the whole mixture was treated in accordance with  
 160 the sample preparation protocol (Fig. 2). The blank was processed in the same way as the tests, replacing the KI with  
 161 ultrapure water.

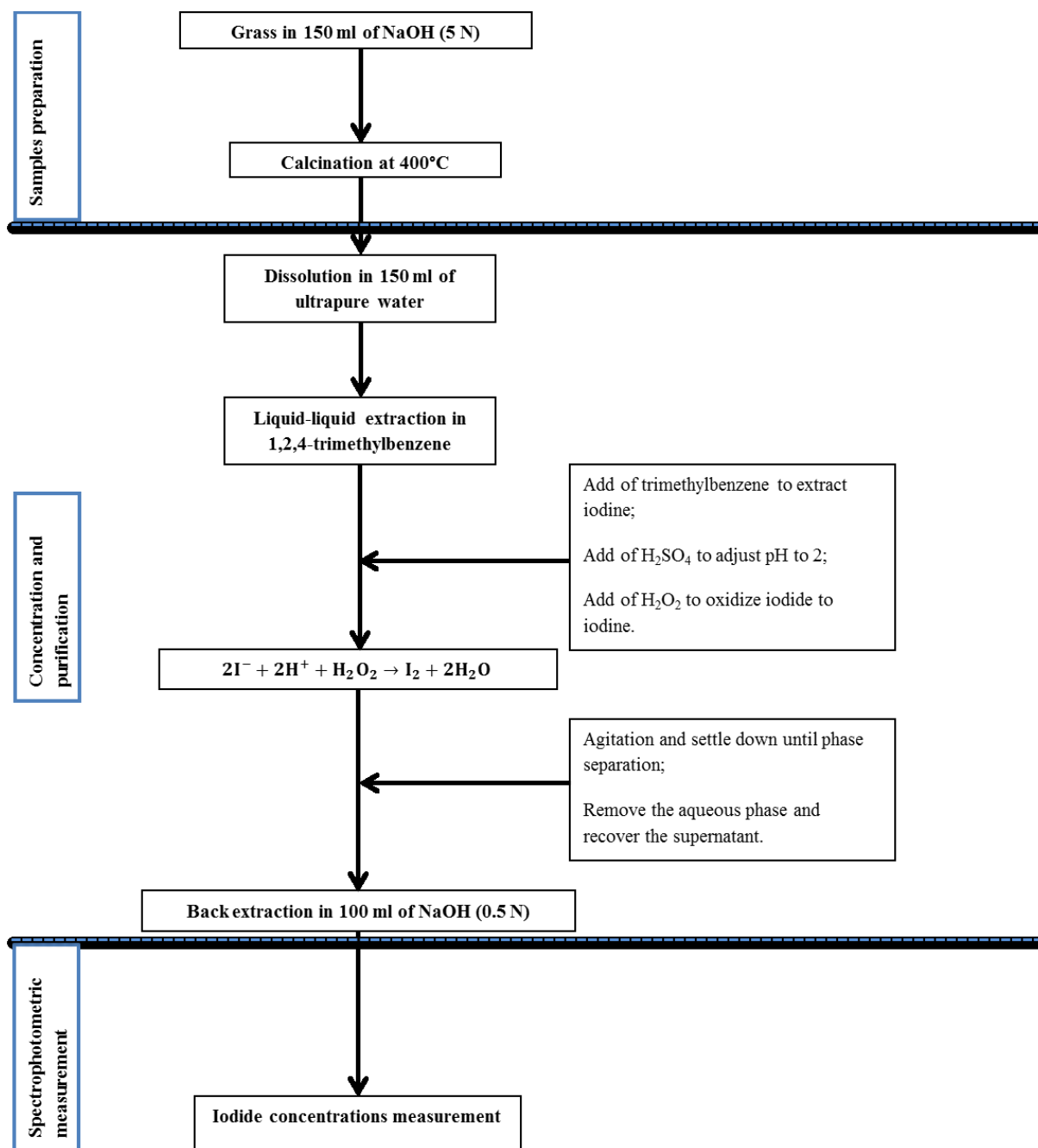


Fig. 2. Processing of grass samples for spectrophotometric measurement.

### 5. Iodine measurement by colorimetry

All the samples were measured by UV-visible spectrophotometry (Safas, UVmc1). The colorimetry technique for iodine measurement is based on the Sandell and Kolthoff (1937) method; the reaction between the cerium ions and the arsenic ions is catalysed by the iodide ions. The absorption measured by spectrophotometry at 415 nm is then inversely



169 proportional to the iodide concentration. The detection limit is  $2.5 \mu\text{g.L}^{-1}$ . The iodine concentrations measured in the  
170 atmosphere are expressed in  $\mu\text{g.m}^{-3}$  and those measured on the grass in  $\mu\text{g.m}^{-2}$ .

## 171 *B. Environmental (including meteorological) variable measurements*

172 An ultrasonic anemometer (Young 81000V, Inc.) set to a frequency of 10 Hz and installed 50 cm above the ground,  
173 and a weather station (Spectrum, Watchdog 2700, Inc.) were used to acquire the meteorological data. The direction ( $^{\circ}$ ),  
174 velocity (u) and friction velocity ( $u^*$ ) of the wind, sensible heat flux (H), Monin-Obukhov length (L) and atmospheric  
175 stability ( $1/L$ ) were measured by the ultrasonic anemometer, and the temperature ( $T_s$ ), relative humidity (RH), total solar  
176 radiation (SR) and photosynthetically active radiation (PAR) were measured by the weather station.

## 177 *C. Modelling of the dry deposition velocities of iodine*

### 178 *1. Model description and identification of the input variables*

179 The dry deposition velocity of a gas refers to the transfer of that gas from the air to surfaces (soil, water, vegetation)  
180 (Arya, 1999) in the absence of precipitation. It is complex and varies significantly according to:

- 181     ▪ the micrometeorological characteristics of the atmospheric surface layer because of the turbulent transport that can  
182         occur in that layer;
- 183     ▪ the physical and chemical properties of the diffusing species;
- 184     ▪ the physical and chemical properties of the surface cover.

185 The dry deposition velocity of a gas has been defined as the inverse of the sum of the three types of resistance to the  
186 dry deposition of that gas, namely the aerodynamic resistance, the laminar sublayer resistance and the canopy resistance or  
187 surface resistance. The expression used to determine  $V_d$  (Eq. (3)) was given by Seinfeld (1985).

$$V_d = \frac{1}{R_a + R_b + R_c} \quad (3)$$

188 Where  $V_d$  is the dry deposition velocity ( $\text{m.s}^{-1}$ );  $R_a$  is the aerodynamic resistance ( $\text{s.m}^{-1}$ );  $R_b$  is the laminar sublayer  
189 resistance ( $\text{s.m}^{-1}$ ) and  $R_c$  is the canopy resistance or surface resistance ( $\text{s.m}^{-1}$ ).

190 The input parameters (Table 3) used to model the deposition velocities were the parameters related to the physico-chemical  
191 properties of the iodine, the surface properties of the grass and the sampling conditions, i.e.:

- 192     ▪ Reference values: diameter of an iodine molecule ( $D_p$ ), canopy aerodynamic resistance ( $R_{ac0}$ ), ground resistance  
193         ( $R_{g0}$ ), cuticle resistance ( $R_{cut0}$ ) and mesophyll resistance ( $R_m$ );
  - 194     ▪ Variables: leaf area index (LAI), roughness length of the grass ( $z_0$ ), measurement height (z) and minimum stomatal  
195         resistance ( $r_i$ ).
- 196  
197  
198

Table 3: Input parameters of the model for dry deposition of iodine.

Campaign (Date)	D <sub>p</sub> (m)	z (m)	z <sub>0</sub> (m)	LAI	ri (s.m <sup>-1</sup> )	R <sub>m</sub> (s.m <sup>-1</sup> )	R <sub>ac0</sub> (s.m <sup>-1</sup> )	R <sub>g0</sub> (s.m <sup>-1</sup> )	R <sub>cut0</sub> (s.m <sup>-1</sup> )
MIOSEC 2 (18 to 26 September 2018)	2.8×10 <sup>-8</sup>	0.26	0.01	1.5	9999	0	1000	100	50
MIOSEC 3 (4 to 7 June 2019)	2.8×10 <sup>-8</sup>	0.26	0.01	1.5	60	0	1000	100	50

## 2. Determination of the resistances (R<sub>a</sub>, R<sub>b</sub> and R<sub>c</sub>) and the input variables of the model

Aerodynamic resistance refers to the aerodynamic component of transfer governed by microturbulence. It therefore corresponds to the resistance to vertical transfer to the surfaces in the immediate vicinity of the elements (gas, aerosols) through the atmospheric surface layer. The lower limit of the atmospheric surface layer is the roughness length. Of the micrometeorological parameters that influence aerodynamic resistance, wind friction velocity and atmospheric stability are the most important (Arya, 1999). The expressions for R<sub>a</sub> and R<sub>b</sub> shown respectively in Eqs. (4) and (5) have been defined by Padro et al. (1991).

$$R_a = \frac{1}{k u_*} \left[ 0.74 \ln \left( \frac{z}{z_0} \right) - \Psi_H \right]$$

With,

$$\Psi_H = \begin{cases} -4.7 \frac{z}{L} & 0 < \frac{z}{L} < 1 \text{ (stable condition)} \\ 2 \cdot 0.74 \ln \left[ \frac{(1+y)}{2} \right], & y = \left( 1 - 9 \frac{z}{L} \right)^{\frac{1}{2}} & -1 < \frac{z}{L} < 0 \text{ (unstable condition)} \end{cases} \quad (4)$$

Where u\* is the wind friction velocity (ms<sup>-1</sup>); k is the Von Karman constant (k=0.4); z is the reference height at which the deposition velocity is evaluated, i.e. the height from the ground at which the air is sampled; z<sub>0</sub> is the roughness length; Ψ<sub>H</sub> is the stability correction function for heat and L is the Monin-Obukhov length.

During the 2 measurement campaigns, the roughness length of the grass and the air sampling height were 0.01 m and 0.26 m respectively. Depending of Pasquill stability classes, the experiments were carried out under stable, unstable and neutral atmospheric conditions (see Table 7 and Table 8). Equation (4) was applied according to the stable case and unstable case defined by author. In neutral condition, Ψ<sub>H</sub>=1 (Seinfeld and Pandis, 2016) was used for R<sub>a</sub> calculation.

Laminar sublayer resistance (R<sub>b</sub>) is the resistance to diffusion of the elements through the molecular sublayer in direct contact with the surface. R<sub>b</sub> depends on both turbulence and molecular diffusion (Eq. (5)).

$$R_b = \frac{z}{k u_*} \left( \frac{v}{D_i} \right)^{\frac{2}{3}} \quad (5)$$

217 Where  $v$  is the kinematic viscosity of the air ( $v = 1.5 \times 10^{-5} \text{ m}^2.\text{s}^{-1}$ ) and  $D_i$  is the molecular diffusivity (Eq. (6)) of a species  $i$   
 218 in the air (Seinfeld, 1985).  
 219 With,

$$D_i = \frac{k_B T Cu}{6\pi \mu D_p} \quad (6)$$

220 Where  $K_B$  is the Boltzmann constant ( $K_B = 1.38 \times 10^{-23} \text{ K.J}^{-1}$ );  $T$  is the temperature (K);  $Cu$  is the Cunningham correction  
 221 factor for small particles or molecules with a diameter of less than  $1 \mu\text{m}$ ;  $\mu$  is the dynamic viscosity coefficient of air ( $1.8 \times$   
 222  $10^{-5} \text{ kg.m}^{-1}.\text{s}^{-1}$ );  $D_p$  is the diameter of the gas molecule studied. The diameter of a gaseous iodine molecule ( $I_2$ ) is  $2.8 \times 10^{-10}$   
 223 m (Forsythe, 1956). The Cunningham correction (Eq. (7)) was given by Seinfeld (1985).  $\lambda$  is the mean free path of a  
 224 molecule. At ambient pressure and temperature:  $\lambda = 6.8 \times 10^{-8} \text{ m}$ .

$$Cu = 1 + \frac{\lambda}{D_p} \left( 2.54 + 0.8 \exp \left( -\frac{0.55 D_p}{\lambda} \right) \right) \quad (7)$$

225 The overall resistance on the surface of the cover ( $R_c$ ), called the "surface resistance" or "canopy resistance",  
 226 includes the stomatal resistance and the non-stomatal resistance. For surfaces with plant cover, the model used most  
 227 frequently to describe  $R_c$  is the model defined by Zhang et al. (2002b).  $R_c$  was determined using Eq. (8) as defined in Zhang  
 228 et al. (2002a).

$$\frac{1}{R_c} = \frac{1 - W_{st}}{R_{st} + R_m} + \frac{1}{R_{ns}} \quad (8)$$

229 Where  $R_c$  is the surface resistance ( $\text{s.m}^{-1}$ );  $R_{st}$  ( $\text{s.m}^{-1}$ ) is the stomatal resistance;  $R_m$  ( $\text{s.m}^{-1}$ ) is the mesophyll resistance;  $R_{ns}$   
 230 ( $\text{s.m}^{-1}$ ) is the non-stomatal resistance and  $W_{st}$  is the stomatal blocking fraction (no unit).

231 The stomatal resistance is the resistance to opening of the stomata. In other words, it is the resistance to gas  
 232 absorption by the blades of grass. It is the result of biological processes on the surface of the plant cover. Consequently, the  
 233 properties of the grass are very important to the final stage of the deposition process. The parameters influencing stomatal  
 234 resistance are (Arya, 1999):

- 235 - the properties of the plant cover, including the water potential, roughness and leaf area index (LAI). The LAI is the  
 236 area developed by the blades of grass per unit of ground surface area. It is a value that has no physical dimension.  
 237 It was determined using an LAI-meter (LICOR, LAI-2000);
- 238 - the physico-chemical properties of the gas, including solubility and molecular diffusivity;
- 239 - the weather conditions, particularly the vapour-pressure deficit, temperature and solar radiation.

240 The stomatal resistance was calculated using an empirical formula (Eq. (9)) developed by (Wesely, 1989).

$$R_{st} = r_i \{ 1 + [200(SR + 0,1)^{-1}]^2 \} \{ 400[T_s(40 - T_s)]^{-1} \} \quad (9)$$

241 Where  $SR$  is the total solar radiation in ( $\text{W.m}^{-2}$ );  $T_s$  is the air temperature ( $^{\circ}\text{C}$ ) between  $0$  and  $40^{\circ}\text{C}$  and  $r_i$  ( $\text{s.m}^{-1}$ ) is the  
 242 minimum stomatal resistance for water vapour. The last of these varies according to the type of plant cover, the season but

also the chemical species. The meteorological data used for the parameterisation of  $R_{st}$  were obtained using the weather station and correspond to the experiment periods (Table 7 and Table 8). Depending of seasons and landuse type, as urban land, deciduous forest or grassland, Wesely (1989) have defined the values of  $r_i$ . The values of  $r_i$  corresponding to grassland are given in Table 4. Given that MIOSEC 2 and MIOSEC 3 ran from 18 to 26 September 2018 and from 4 to 7 June 2019, the values  $r_i = 9999 \text{ (s.m}^{-1}\text{)}$  and  $r_i = 60 \text{ (s.m}^{-1}\text{)}$  respectively were selected for the calculation of  $R_{st}$ .

**Table 4:**  $r_i \text{ (s.m}^{-1}\text{)}$  values for grassland according to season (Wesely, 1989).

Period of the year	$r_i \text{ (s.m}^{-1}\text{)}$
Summer	60
Autumn before harvest	9999
Late autumn with frost but no snow	9999
Winter with snow	9999
Spring	120

Equation. (10) corresponds to the expression of the stomatal blocking fraction for a wet canopy (Brook et al., 1999). For dry canopy,  $W_{st}$  always equals 0.  $W_{st}$  is given a value other than 0 only when solar radiation is relatively strong ( $> 200 \text{ W.m}^{-2}$ ) and the canopy is wet. If rain or dew occurs, the canopy is treated as wet (Zhang et al., 2003b). In this study, as the experiments were carried out outside of dew and rain periods, the value of 0 was assigned to  $W_{st}$ .

$$W_{st} = \begin{cases} 0 & SR < 200 \text{ (W.m}^{-2}\text{)} \\ \frac{SR - 200}{800} & 200 < SR < 600 \text{ (W.m}^{-2}\text{)} \\ 0.5 & SR > 600 \text{ (W.m}^{-2}\text{)} \end{cases} \quad (10)$$

Mesophyll resistance ( $R_m$ ) is resistance to the diffusion of gas in the mesophyll, preventing it from reaching the chloroplasts (Davi, 2004).  $R_m$  depends specifically on the chemical species. For chemical species with very high solubility and/or high oxidative power,  $R_m$  is negligible, whereas for chemical species with very low solubility and/or low oxidative power,  $R_m$  is assigned a value of  $100 \text{ s.m}^{-1}$ . For all other chemical species,  $R_m$  is 0 (Zhang et al., 2002b). In view of both the very low solubility of iodine ( $I_2$ ) in water (Gottardi, 2001) and its high oxidative power (Carpenter, 2003), a value of 0 has been assigned to  $R_m$ .

The expression for non-stomatal resistance (Eq. (11)) was the expression determined when parameterising  $R_{ns}$  for  $O_3$  (Zhang et al., 2002a).

$$\frac{1}{R_{ns}} = \frac{1}{R_{ac} + R_g} + \frac{1}{R_{cut}} \quad (11)$$

Where  $R_{ns}$  is the non-stomatal resistance ( $\text{s.m}^{-1}$ );  $R_{ac} \text{ (s.m}^{-1}\text{)}$  is the canopy aerodynamic resistance;  $R_g \text{ (s.m}^{-1}\text{)}$  is the ground resistance and  $R_{cut} \text{ (s.m}^{-1}\text{)}$  is the cuticle resistance.  $R_{ac}$  does not depend on the chemical species whereas  $R_g$  and  $R_{cut}$  do depend on it.

$R_{ac}$  is affected by the change in canopy structure including LAI and wind friction velocity.  $R_{ac}$  expression (Eq. (12)) is given by Zhang et al. (2002b).

$$R_{ac} = \frac{R_{ac0} \times LAI^{1/4}}{u_*^2} \quad (12)$$

Where  $R_{ac0}$  ( $s.m^{-1}$ ) is the reference value for  $R_{ac}$  ( $s.m^{-1}$ ); LAI is leaf area index and  $u^*$  is the wind friction velocity ( $m.s^{-1}$ ).  $R_{ac0}$  values are determined and assigned at many type of canopy including grass. The value of  $R_{ac0}$  ( $50 s.m^{-1}$ ) used in the model corresponds to that defined by Wesely (1989) for grassland.  $R_g$  and  $R_{cut}$  were calculated for  $SO_2$  and  $O_3$ . For all other gases, a formulation to determine  $R_g$  and  $R_{cut}$  (Eq. (13)) was developed by Zhang et al, (2003).

$$\frac{1}{R_x(i)} = \frac{\alpha(i)}{R_x(SO_2)} + \frac{\beta(i)}{R_x(O_3)} \quad (13)$$

Where  $R_x$  is the ground resistance ( $R_g$ ) or cuticular resistance ( $R_{cut}$ );  $i$  represent the chemical species;  $\alpha$  and  $\beta$  are 2 factors based on solubility and semi-redox reactivity of  $i$  (Wesely, 1989). Previous studies have suggested that  $R_c$  of iodine would be identical to that of  $SO_2$  (Brandt et al., 2002). As the parameters  $\alpha$  and  $\beta$  for iodine are not known, approximations have been made. Iodine like  $SO_2$  has high semi-redox reactivity. Although the solubility in water at  $25^\circ C$  of iodine ( $0.34 g.L^{-1}$ ) (Hartley and Campbell, 1908) is much lower than that of  $SO_2$  ( $82.4 g.L^{-1}$ ) (Byerley et al., 1980), as long as the quantity of iodine emitted in atmosphere can be dissolved and oxidized on the grass surface, solubility and semi-redox reactivity may be considered as not a limiting factors and, consequently that parameters  $R_g$  and  $R_{cut}$  of  $SO_2$  can be used for  $I_2$ .  $R_g$  is considered depending of surface type. For different chemical species including  $I_2$ ,  $R_g$  would be different depending that canopy is dry or wet (dew or rain). For a dry canopy,

$$R_{cut} = \frac{R_{cutd0}}{e^{0.03 \times RH} \times LAI^{1/4} \times u_*} \quad (14)$$

For a wet canopy,

$$R_{cut} = \frac{R_{cutw0}}{LAI^{1/2} \times u_*} \quad (15)$$

Where  $R_{cutd0}$  ( $s.m^{-1}$ ) and  $R_{cutw0}$  ( $s.m^{-1}$ ) represent the reference values of  $R_{cut}$  ( $s.m^{-1}$ ) for dry and wet canopy, respectively; RH is relative humidity (%); LAI is the leaf area index and  $u^*$  is the wind friction velocity ( $m.s^{-1}$ ). The reference values  $R_{g0}$  ( $100 m.s^{-1}$ ) used in this study is that for  $SO_2$  for grass (Zhang et al., 2003a). As the experiments were carried out outside of dew and rain periods, the reference value  $R_{cutd0}$  ( $1000 s.m^{-1}$ ) used is that for  $SO_2$  for grass in dry conditions (Zhang et al., 2003a). Equations. (16) and (17) are the resultants of the integration of the expressions for  $R_{ac}$ ,  $R_g$  and  $R_{cut}$  in Eq. (11). The expression for  $R_{ns}$  becomes: For a dry canopy,

$$\frac{1}{R_{ns}} = \frac{1}{R_{ac0} u^{*-2} LAI^{0.25} + R_{g0}} + \frac{1}{R_{cut0} e^{(-0.03 RH)} LAI^{-0.25} u^{*-1}} \quad (16)$$

For a wet canopy,

$$\frac{1}{R_{ns}} = \frac{1}{R_{ac0} u^{*-2} LAI^{0.25} + R_{g0}} + \frac{1}{R_{cut0} LAI^{-0.5} u^{*-1}} \quad (17)$$

A LAI of 1.50 was used as an input parameter to calculate the non-stomatal resistance. This value was set as a result of the measurements made with the LAI-meter. The meteorological parameters used to model the non-stomatal resistance are given in Table 7 and Table 8.

### 3. Results and discussion

#### *A. Measurement of iodine dry deposition velocities on grass*

The iodine concentrations measured in blanks flasks and filters were below the detection limit (see Appendix 1 and 2). Consequently, there was no transformation of iodine during emission and iodine measured in samples corresponds to that emitted using iodine generator.

To determine the quantity of iodine in grass tests, the quantity of iodine in blank grass is subtracted from the quantity of iodine measured in grass tests. Extraction efficiency is determined by dividing the quantity of iodine measured in grass tests to the quantity of iodine added. This ratio is expressed as a percentage. For the 12 tests performed, the mean extraction efficiency was 57% with a ratio of standard deviation to mean (RSD) of 6.44%. These results validated the technique for separation of the iodine as  $I_2$  form deposited on the grass. The quantity of iodine measured on the grass blank was  $0.14 \pm 0.01 \mu\text{g.g}^{-1}$  of fresh grass (mass  $\pm$  RSD for the extraction efficiency applied to the mass). The application of this technique to the samples resulting from the iodine emissions determined the iodine dry deposition velocities.

**Table 5:** Dry deposition velocities of gaseous elemental iodine ( $^{127}\text{I}$ ) on grass during MIOSEC 2.

Date and time	Atmospheric iodine concentration in $\mu\text{g.m}^{-3}$	Dry deposition flux of iodine in $\mu\text{g.m}^{-2}.\text{s}^{-1}$	Dry deposition velocity of iodine in $\text{cm.s}^{-1}$
19/09/2018 10:32	110.6	0.078	$0.07 \pm 0.005$
19/09/2018 15:00	29.7	0.076	$0.25 \pm 0.016$
20/09/2018 09:30	18.4	0.010	$0.06 \pm 0.004$
20/09/2018 14:32	48.7	0.063	$0.13 \pm 0.008$
20/09/2018 17:30	30.9	0.039	$0.13 \pm 0.008$
21/09/2018 15:00	46.5	0.046	$0.10 \pm 0.006$
21/09/2018 18:30	116.7	0.024	$0.02 \pm 0.001$
22/09/2018 09:30	117.6	0.081	$0.07 \pm 0.005$
22/09/2018 17:00	15.2	0.042	$0.28 \pm 0.018$
24/09/2018 10:30	20.9	0.058	$0.28 \pm 0.018$
24/09/2018 16:30	16.1	0.079	$0.49 \pm 0.032$
25/09/2018 09:30	4.7	0.012	$0.25 \pm 0.016$
26/09/2018 10:00	4.9	0.002	$0.04 \pm 0.003$

27/09/2018 10:30	132.9	0.399	0.30 ± 0.019
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The dry deposition velocities measured during MIOSEC 2 (Table 5) are between 0.02 and 0.49 cm.s<sup>-1</sup>.

**Table 6:** Dry deposition velocities of gaseous elemental iodine (<sup>127</sup>I) on grass during MIOSEC 3.

Date and time	Atmospheric iodine concentration in µg.m <sup>-3</sup>	Dry deposition flux of iodine in µg.m <sup>-2</sup> .s <sup>-1</sup>	Dry deposition velocity of iodine in cm.s <sup>-1</sup>
04/06/2019 16:05	11.8	0.15	1.25 ± 0.08
04/06/2019 17:32	8.8	0.08	0.87 ± 0.06
06/06/2019 07:35	44.6	0.21	0.48 ± 0.03
06/06/2019 10:30	13.2	0.10	0.73 ± 0.05
06/06/2019 12:30	18.9	0.13	0.69 ± 0.04
06/06/2019 16:03	12.3	0.08	0.63 ± 0.04
07/06/2019 09:55	28.2	0.32	1.15 ± 0.07
07/06/2019 10:47	15.3	0.17	1.10 ± 0.07

During MIOSEC 3, the dry deposition velocities measured (Table 6) are between 0.48 and 1.25 cm.s<sup>-1</sup>.

### *B. Environmental (including meteorological) variable measurements*

The meteorological data obtained using the ultrasonic anemometer and the weather station during the experiment periods are presented in Table 7 and Table 8, which correspond to MIOSEC 2 and MIOSEC 3 respectively.

**Table 7:** Meteorological data for the 14 experiments conducted during MIOSEC 2.

Date and time	Ts (°C)	S (W.m <sup>-2</sup> )	H (W.m <sup>-2</sup> )	RH (%)	u* (m.s <sup>-1</sup> )	1/L	Stability
19/09/2018 10:32	21	426	75	71	0.16	-0.268	Unstable
19/09/2018 15:00	26	798	211	39	0.37	-0.059	Unstable
20/09/2018 09:30	18	286	14	97	0.07	-0.584	Unstable
20/09/2018 14:32	27	782	181	36	0.26	-0.145	Unstable
20/09/2018 17:30	28	386	84	36	0.24	-0.089	Unstable
21/09/2018 15:00	20	653	169	47	0.33	-0.069	Unstable
21/09/2018 18:30	17	115	9	47	0.22	-0.012	Neutral
22/09/2018 09:30	12	119	9	95	0.05	-0.826	Unstable
22/09/2018 17:00	22	475	83	31	0.35	-0.029	Unstable
24/09/2018 10:30	12	433	71	71	0.36	-0.022	Unstable
24/09/2018 16:30	18	542	165	37	0.53	-0.016	Neutral
25/09/2018 09:30	10	272	27	66	0.36	-0.008	Neutral
26/09/2018 10:00	12	340	38	57	0.15	-0.169	Unstable
27/09/2018 10:30	19	435	45	45	0.26	-0.038	Unstable

**Table 8:** Meteorological data for the 8 experiments conducted during MIOSEC 3.

Date and time	Ts (°C)	S (W.m <sup>-2</sup> )	H (W.m <sup>-2</sup> )	RH (%)	u* (m.s <sup>-1</sup> )	1/L	Stability
04/06/2019 16:05	20	216	47	63	0.47	-0.005	Neutral
04/06/2019 17:32	19	293	39	69	0.31	-0.018	Neutral
06/06/2019 07:35	9	58	6	100	0.12	0.027	Stable
06/06/2019 10:30	13	344	59	84	0.23	-0.068	Unstable
06/06/2019 12:30	16	644	89	60	0.24	-0.090	Unstable
06/06/2019 16:03	19	725	94	46	0.33	-0.038	Unstable

07/06/2019 09:55	12	180	17	95	0.43	-0.001	Neutral
07/06/2019 10:47	14	389	83	86	0.47	-0.011	Neutral

### C. Modelling of the dry deposition velocities of iodine

The iodine dry deposition velocities obtained by the model are between 0.11  $\text{cm.s}^{-1}$  and 0.51  $\text{cm.s}^{-1}$  during MIOSEC 2 (Table 9) and between 0.31  $\text{cm.s}^{-1}$  and 1.60  $\text{cm.s}^{-1}$  during MIOSEC 3 (Table 10).

**Table 9:** Dry deposition velocities of iodine produced by the model during MIOSEC 2.

Date and time	$R_a (\text{s.m}^{-1})$	$R_b (\text{s.m}^{-1})$	$R_{st} (\text{s.m}^{-1})$	$R_c (\text{s.m}^{-1})$	$V_d (\text{cm.s}^{-1})$
19/09/2018 10:32	11	2	12219	505	0.19
19/09/2018 15:00	6	1	11734	294	0.33
20/09/2018 09:30	19	6	14988	641	0.15
20/09/2018 14:32	8	1	11953	492	0.20
20/09/2018 17:30	9	2	14923	562	0.17
21/09/2018 15:00	7	1	10937	316	0.31
21/09/2018 18:30	16	2	40963	533	0.18
22/09/2018 09:30	20	7	45588	903	0.11
22/09/2018 17:00	6	1	11844	356	0.28
24/09/2018 10:30	6	1	14803	188	0.51
24/09/2018 16:30	7	1	11519	193	0.50
25/09/2018 09:30	10	1	20977	206	0.46
26/09/2018 10:00	13	3	15917	749	0.13
27/09/2018 10:30	9	1	12169	453	0.22

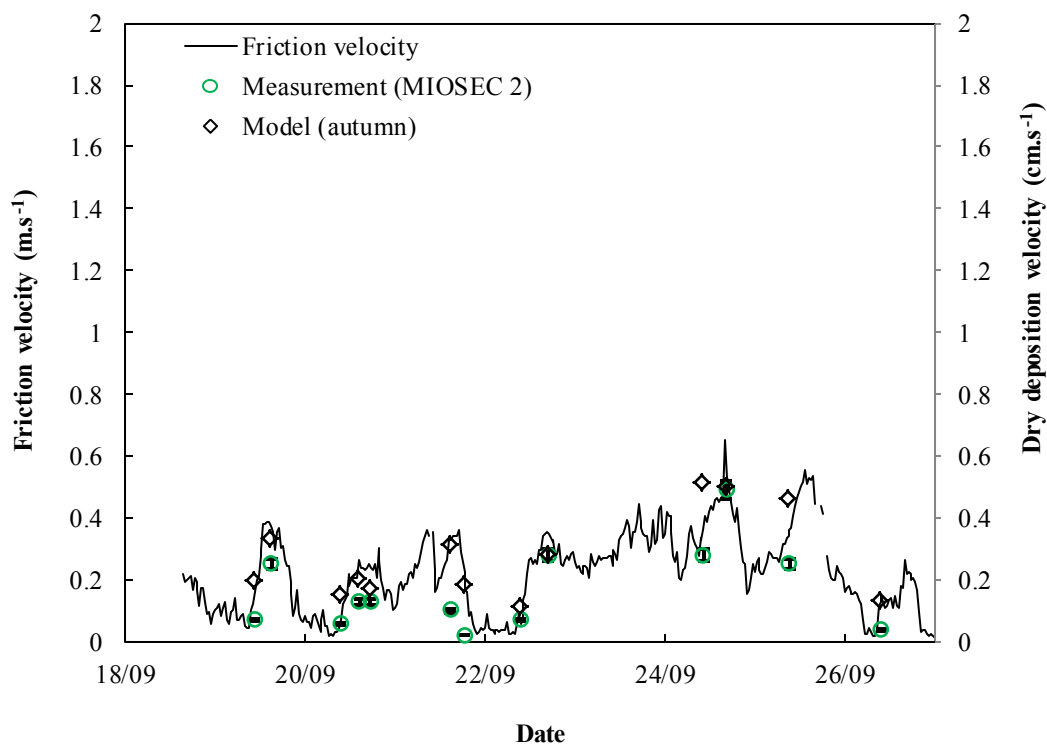
**Table 10:** Dry deposition velocities of iodine produced by the model during MIOSEC 3.

Date and time	$R_a (\text{s.m}^{-1})$	$R_b (\text{s.m}^{-1})$	$R_{st} (\text{s.m}^{-1})$	$R_c (\text{s.m}^{-1})$	$V_d (\text{cm.s}^{-1})$
04/06/2019 16:05	8	2	111	66	1.32
04/06/2019 17:32	11	3	88	71	1.17
06/06/2019 07:35	51	7	1108	263	0.31
06/06/2019 10:30	10	4	90	76	1.12
06/06/2019 12:30	9	5	68	101	0.87
06/06/2019 16:03	7	4	65	93	0.96
07/06/2019 09:55	8	2	159	59	1.45
07/06/2019 10:47	8	2	84	53	1.60



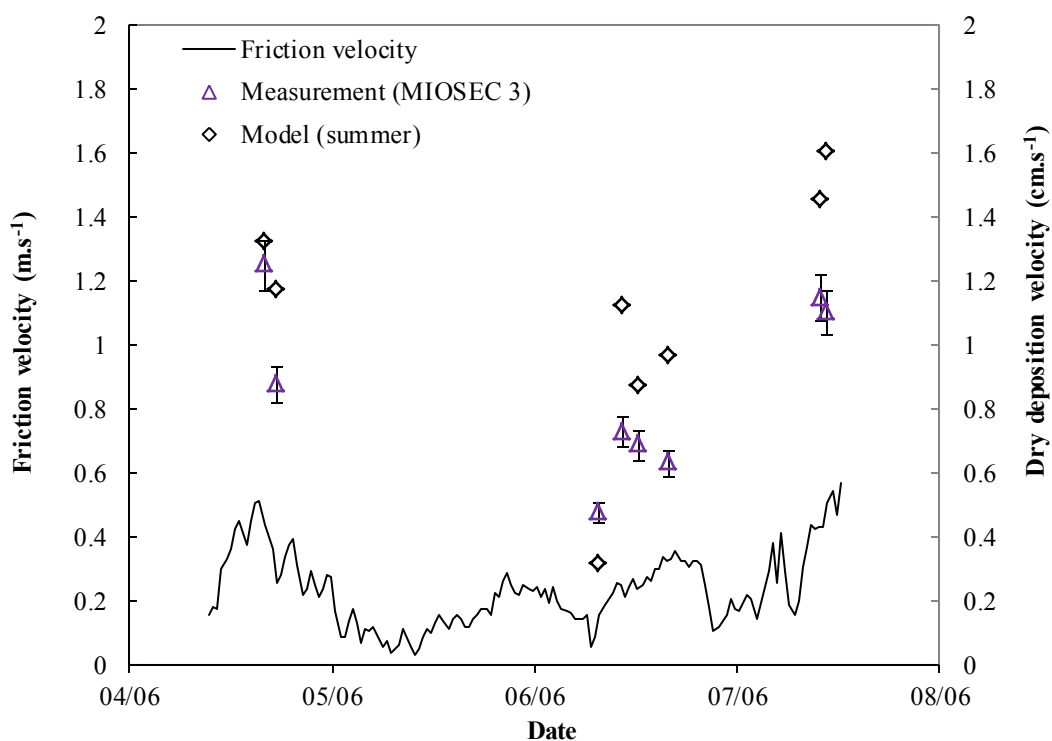
333 *D. Change in the dry deposition velocities of iodine as a function of environmental variables and the*  
 334 *physiological properties of the grass*

335 During MIOSEC 2, the deposition velocities varied by more than on order of magnitude and values produced by the  
 336 model varied as the same of measurements results. Although the data is very limit on statistical significance, the reasons to  
 337 affect the dry deposition velocity during MIOSEC 2 are the mechanical turbulence and, more specifically, in the wind  
 338 friction velocity (Fig. 3). These differences are due to the variations existing between the wind frictions velocities  
 339 (approximately on order of magnitude) measured at the same moment of experiments. Deposition velocities are higher  
 340 during the periods of strong winds than during periods of light winds. This observation is corroborated by the significance  
 341 level of the correlation (Fig. 5) of the modelled deposition velocities ( $V_{d_{mod}}$ ) with the wind friction velocities ( $u^*$ ) ( $V_{d_{mod}} =$   
 342  $1.0 \times u^*$ ,  $R^2 = 0.7$ ). In other words, the iodine deposition on the grass due to entrainment of iodine onto the surface of the  
 343 blades of grass by the wind friction velocity is related to surface area. Unlike wind friction velocity, the correlation of  
 344 deposition velocities with sensible heat flux and stomatal resistance is not significant. Furthermore, in view of the stomatal  
 345 resistances ( $R_{st}$ ) determined during MIOSEC 2, the model shows that the experiments were conducted during a period of the  
 346 year (autumn) when the stomata were only slightly open. Consequently, the deposition velocities were dependent on the  
 347 non-stomatal resistance ( $R_{ns}$ ), which is also dependent on the wind friction velocity.



348  
 349 Fig. 3. Change in dry deposition velocities of gaseous elemental iodine and wind friction velocities during  
 350 MIOSEC 2: the bar on the deposition velocities (circles) corresponds to the mean standard deviation divided by the mean as  
 351 a percentage for the extraction efficiency (RSD = 6.34%).  
 352

353 During MIOSEC 3, the variations in measurements deposition velocities are the same to values given by model. The first  
 354 reason of variations in deposition velocities is due to the mechanical turbulence. Deposition velocities are higher during  
 355 periods of strong winds than during periods of light winds. The second reason is due to the stomata opening. For the same  
 356 values of friction velocities, dry deposition velocities obtained during MIOSEC 3 are 2 to 3 times higher than those obtained  
 357 during MIOSEC 2. The model shows that the stomatal resistances were smaller during MIOSEC 3 than those obtained  
 358 during MIOSEC 2. In other words, the experiments were conducted during a period of the year (summer) when the stomata  
 359 were more open. Consequently, the dry deposition velocities were higher in this period. The change in deposition velocity  
 360 during MIOSEC 3 shows that the dry deposition of iodine was favoured not only by the wind friction velocity (Fig. 4) but  
 361 also by iodine penetration into the blades of grass due to the openness of the stomata. This is reflected in the high  
 362 significance level of the correlation (Fig. 5) of the modelled deposition velocities with the wind friction velocities ( $Vd_{mod} =$   
 363  $3.0 \times u^*$ ,  $R^2 = 0.9$ ).



364  
 365 Fig. 4. Change in dry deposition velocities of gaseous elemental iodine and wind friction velocities during MIOSEC 3: the  
 366 bar on the deposition velocities (triangles) corresponds to the mean standard deviation divided by the mean as a percentage  
 367 for the extraction efficiency (RSD = 6.34%).

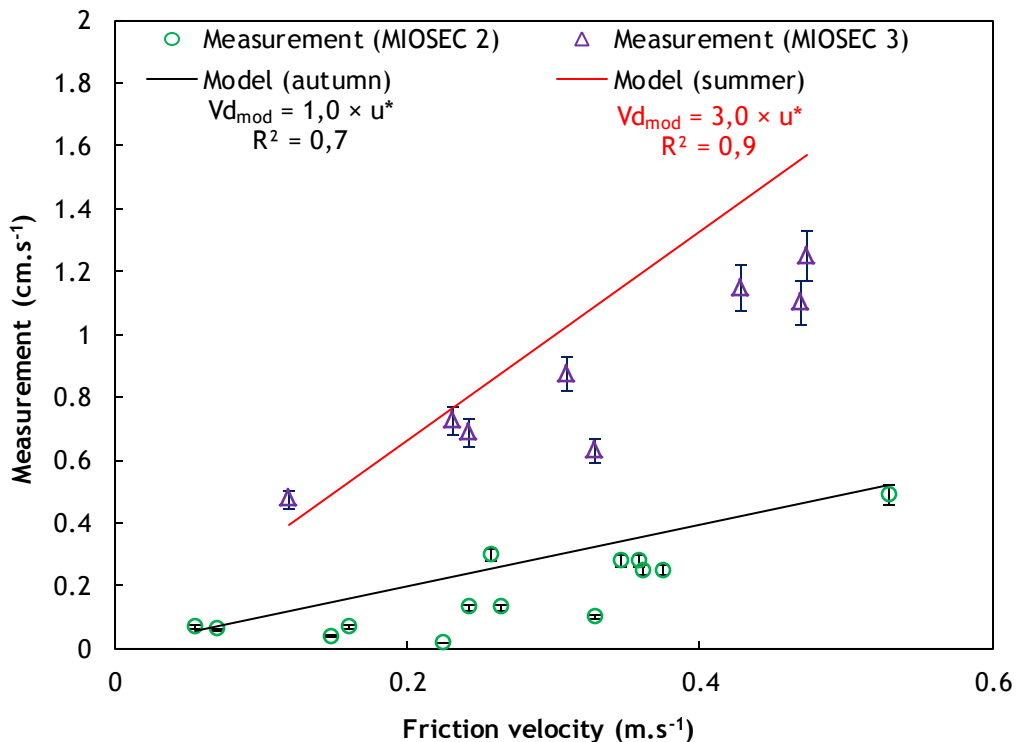


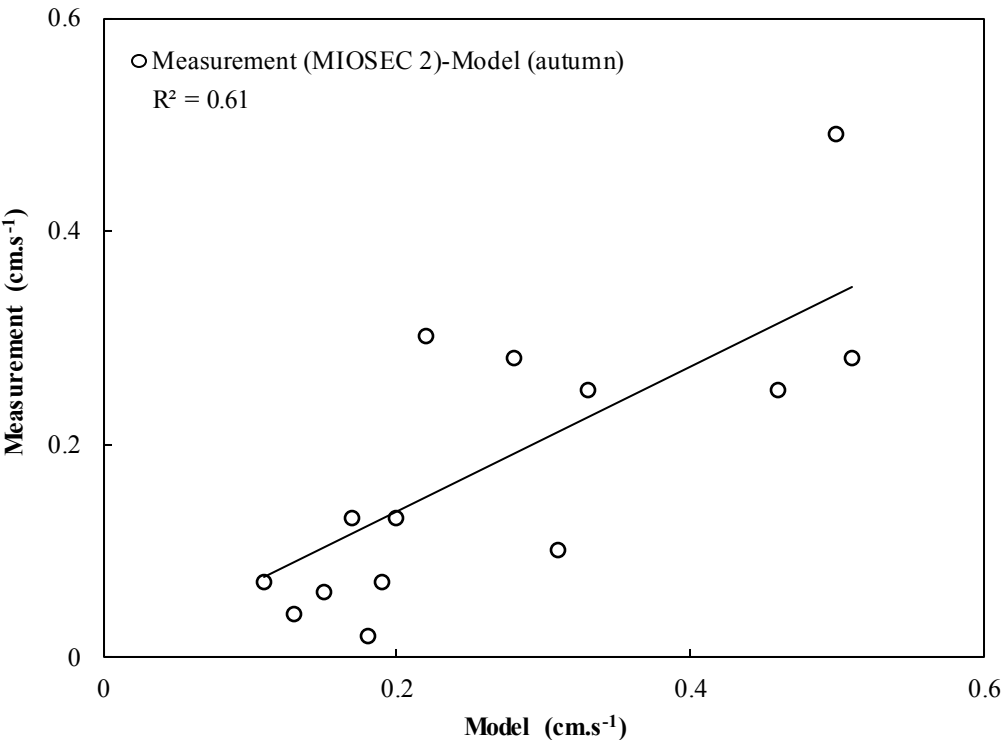
Fig. 5. Change in dry deposition velocities of gaseous elemental iodine during MIOSEC 2 (circles) and MIOSEC 3 (triangles) as a function of the wind friction velocities: the bar on the deposition velocities corresponds to the mean standard deviation divided by the mean as a percentage for the extraction efficiency (RSD = 6.34%).

#### E. Comparison of the measured dry deposition velocities with the modelled dry deposition velocities

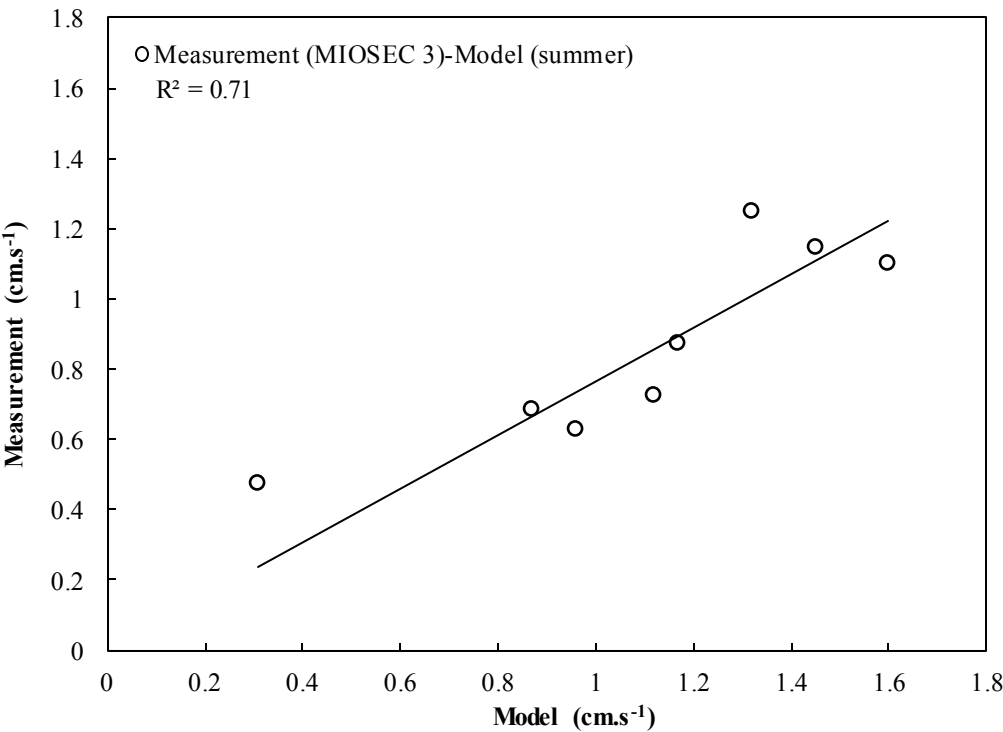
The measured dry deposition velocities are comparable with the modelled dry deposition velocities and, when the former are related by a function to the latter for each of the measurement campaigns, they correlate linearly with correlation coefficients ( $R^2$ ) of 0.61 and 0.71 for MIOSEC 2 (Fig. 6) and MIOSEC 3 (Fig. 7), respectively. The measured data are generally 15-30% lower than those modelling results during both MIOSEC 2 and 3. The average of the deviations in absolute value compared to the measured values (Mean Absolute Percentage Error, MAPE) is 10% and 5% during MIOSEC 2 and 3, respectively. Knowing that the solubility of  $\text{SO}_2$  is higher than that  $\text{I}_2$  and  $R_{g0}$  and  $R_{cut0}$  being parameters based on solubility and semi-redox reactivity of the gaseous specie, the overestimated results of model could be explained by the fact that approximations consisting to apply values references of  $R_{g0}$  and  $R_{cut0}$  from  $\text{SO}_2$  to  $\text{I}_2$  were used in the model in place of the reference values specific to  $\text{I}_2$ . The model can be improved as a determining the parameters  $R_{g0}$  and  $R_{cut0}$  specific to  $\text{I}_2$ .

To conclude, the modelled dry deposition velocities and the measured dry deposition velocities show that the dry deposition of iodine is dependent on surface resistance ( $R_c$ ), which is depends on stomatal resistance ( $R_{st}$ ) and non-stomatal resistance ( $R_{ns}$ ). The model also shows that stomatal resistance is lower in summer than in autumn, whereas non-stomatal resistance varies very slightly through the seasons and is dependent on the wind friction velocity ( $u^*$ ). Consequently, when stomatal resistance is high, which coincides with closure of the stomata, dry deposition velocities are lower and are

388 dependent on the wind friction velocities. When stomatal resistance falls, which coincides with opening of the stomata, the  
 389 deposition velocities increase and depend on both the wind friction velocities and the openness of the stomata.



390  
 391 **Fig. 6.** Comparison of measured dry deposition velocities with modelled dry deposition velocities during MIOSEC 2.



392  
 393 **Fig. 7.** Comparison of measured dry deposition velocities with modelled dry deposition velocities during MIOSEC 3.

## 394 *F. Comparison with the data in the literature*

395 Numerous data related to determination of the dry deposition velocities of iodine exist in the literature (Table 11).  
396 These values are by no means exhaustive but only examples. The deposition velocities of  $I_2$  has been measured to a range of  
397 different surfaces both in the outdoor and indoor environment including skin and relate to both stable iodine ( $^{127}I$ ) and  
398 radioactive iodine ( $^{129}I$ ,  $^{131}I$ ) in their elemental or particulate forms.

399 It is mentioned in table 11 that certain dry deposition velocities of iodine corresponded to those iodine in particulate  
400 form, it should be remember here the reasons of the differences in these deposition with those of elementary iodine  
401 measured in this study. These differences are due to the fact that the diffusivity of gases is greater than that of particles.

402 For experiments conducted in the environment, the data reported showed that dry deposition velocities for iodine  
403 were between 0.001 and 1  $\text{cm.s}^{-1}$  (Handge and Hoffman, 1974), which means that the data were spread across three orders  
404 of magnitude. The dry deposition velocities measured in that study are similar to those reported here, though the data  
405 dispersion is significantly smaller for the data acquired during MIOSEC 3 (0.48-1.25  $\text{cm.s}^{-1}$ ) and is limited to one order of  
406 magnitude in MIOSEC 2 (0.02-0.49  $\text{cm.s}^{-1}$ ). Concerning the dry deposition velocities of gaseous elemental iodine, the  
407 measurements presented by many studies are spread across several orders of magnitude. For example, measurements taken  
408 on grass by Chamberlain and Chadwick (1966) led to the conclusion that the dry deposition velocity of elemental iodine  
409 ( $^{131}I$ ) was 1.8  $\text{cm.s}^{-1}$ . Most authors suggest taking 1  $\text{cm.s}^{-1}$  as the dry deposition velocity of elemental iodine, though  
410 Heinemann and Vogt (1980) suggest 2  $\text{cm.s}^{-1}$ . The mean of the deposition velocities measured in this study during  
411 MIOSEC 3 (0.86  $\text{cm.s}^{-1}$ ) is similar to the value suggested in the other studies (1  $\text{cm.s}^{-1}$ ). For MIOSEC 2, two main reasons  
412 could explain the difference between these suggested values and the values obtained.

413 The first reason could be the time of day when the dry deposition velocities were measured and the variations in the  
414 meteorological parameters, including wind friction velocity, temperature and solar radiation. During this study, the dry  
415 deposition velocities measured during MIOSEC 2 did indeed vary in the course of the experiments, in line with the  
416 variations in wind friction velocity.

417 The second possible reason could be the period of the year when the measurements were taken. Measurements of  $^{127}I$   
418 have led to the calculation of dry deposition velocities of 0.16  $\text{cm.s}^{-1}$  (Chamberlain and Chadwick, 1966), similar to the  
419 mean of the dry deposition velocities obtained in this study during MIOSEC 2 (0.18  $\text{cm.s}^{-1}$ ). The results obtained in this  
420 study during MIOSEC 2 are also similar to those obtained when determining the dry deposition velocities for  $O_3$  on  
421 grassland at night, i.e. when the stomata were closed, so when the dry deposition velocities were strictly linked to surface  
422 resistance, which is inversely linked to wind friction velocity (Zhang et al., 2002b). According to the model produced  
423 during this study, the dry deposition velocities of iodine depend on the surface resistance ( $R_c$ ), which is a combination of  
424 non-stomatal resistance ( $R_{ns}$ ) and stomatal resistance ( $R_{st}$ ). The model showed that  $R_{ns}$  varies only very slightly through the  
425 seasons of the year and depends on the wind friction velocity ( $u^*$ ). The model also showed that  $R_{st}$  is higher in autumn,  
426 when the stomata are closed, than in summer, when the stomata are open. For example, the deposition velocities measured  
427 in this study were higher in summer (MIOSEC 3) than in autumn (MIOSEC 2). The mean surface resistance given by the  
428 model was 456  $\text{s.m}^{-1}$  for MIOSEC 2 and 98  $\text{s.m}^{-1}$  for MIOSEC 3. If reference were made to Chamberlain and Chadwick  
429 (1966), who suggested taking  $R_c = 500 \text{ s.m}^{-1}$  when determining the dry deposition velocities of gaseous  $^{131}I$ , then the dry  
430 deposition velocities of iodine would be deemed not to change throughout the seasons, which contradicts our observations.

431 **Table 11:** Dry deposition velocities of iodine reported in the literature.

Dry deposition velocity (cm.s <sup>-1</sup> )	Test specimens	Observations	Reference
10 <sup>-3</sup> -1	Grass	Review, <sup>131</sup> I.	(Handge and Hoffman, 1974)
0.1-1.5	Grass	Elemental and particulate iodine, <sup>131</sup> I.	(Eggleton et al., 1963)
0.1-0.5	Grass	Fallout from Fukushima measured in France, <sup>131</sup> I.	(Parache et al., 2011)
0.54	Grass	Calculation of iodine deposits from data reported for distant nuclear explosions in the British Isles.	(Chamberlain and Chadwick, 1966)
0.5	Grass	Calculation of iodine deposits from data reported for distant nuclear explosions, Brookhaven, Long Island.	(Hull, 1963)
0.58	Grass	Air stack at the Karlsruhe reprocessing plant, <sup>129</sup> I.	(Wershofen and Aumann, 1989)
0.5	Grass	Elemental and particulate iodine, <sup>131</sup> I.	(Hull, 1963)
0.3	Grass	Elemental and particulate iodine, <sup>131</sup> I.	(Clark and Smith, 1988)
0.8	Grass	Elemental iodine, <sup>131</sup> I.	(Chamberlain, 1959)
1.8	Grass	Field experiments, elemental iodine.	(Chamberlain and Chadwick, 1966)
2	Grass	Elemental iodine, <sup>131</sup> I.	(Heinemann and Vogt, 1980)
(0.5 – 3.3) × 10 <sup>-3</sup>	Grass	Environmental chamber, <sup>127</sup> I.	(Karunakara et al., 2018)
5.4 × 10 <sup>-5</sup>	Bean leaf	Environmental chamber, <sup>131</sup> I.	(Singhal et al., 2004)

432  
433 Finally, based on closed field experiments (environmental chamber), with the emission of gaseous <sup>127</sup>I on grass,  
434 Karunakara et al. (2018) propose dry deposition velocities of (0.51-3.3) × 10<sup>-3</sup> cm.s<sup>-1</sup>, whereas with the emission of gaseous  
435 <sup>131</sup>I on bean leaves, Singhal et al. (2004) propose 5.4 × 10<sup>-5</sup> cm.s<sup>-1</sup>. These values differ by several orders of magnitude from  
436 those obtained during this study and from data produced during experiments in the environment. These differences are due  
437 to the fact that, in an open atmosphere, deposition is controlled by atmospheric turbulence.

438

#### 439 4. Conclusion

440 An experimental methodology based on emissions of gaseous elemental iodine (I<sub>2</sub>) into the atmosphere in the middle  
441 of grassland was developed to determine the dry deposition velocities of iodine. The dry deposition velocities measured  
442 during MIOSEC 2 are between 0.02 and 0.50 cm.s<sup>-1</sup>, a range covering one order of magnitude. The dry deposition velocities  
443 measured during the MIOSEC 3 campaign are between 0.48 and 1.25 cm.s<sup>-1</sup>. As a result of these experiments, a model for  
444 the dry deposition of iodine was developed. This model is based on the gas dry deposition principle. The input parameters  
445 for the model were the micrometeorological characteristics of the atmospheric surface layer, the physico-chemical  
446 properties of the iodine and the surface properties of the grass, all measured at the time of the experiments. The deposition  
447 velocities produced by the model are between 0.11 and 0.51 cm.s<sup>-1</sup> for the MIOSEC 2 campaign and between 0.31 and 1.60  
448 cm.s<sup>-1</sup> for the MIOSEC 3 campaign. For both campaigns, the measured dry deposition velocities are comparable with the  
449 modelled dry deposition velocities and when the former are related by a function to the latter, they correlate linearly with  
450 significant correlation coefficients. Consequently, this study qualifies the model's input parameters. The model showed that  
451 the variations between deposition velocities are due to variations in the wind friction velocities measured at the time of the  
452 experiments. The model also showed that the deposition velocities changed through the different seasons of the year. The

deposition velocities are higher in summer, when the openness of the stomata is at its maximum. The deposition velocities measured in summer (MIOSEC 3) depend on both the wind friction velocities and the openness of the stomata. They are lower in autumn when they are mainly dependent on the wind friction velocities because of the closure of the stomata. The data obtained during this study agree with those reported in the literature for experiments conducted under the same conditions. However, they differ from the data obtained during experiments conducted in experimental chambers where there is no atmospheric turbulence. It should be noted that this study consisting of modelling the deposition of I<sub>2</sub> and comparing the results of model with values obtained experimentally is original. To improve model, the approximations made must be replaced by the parameters specific to I<sub>2</sub>. The model is not suitable for atmospheric process for prognostic pollution models, as it requires measurement of a range of parameters. However, it remains applicable to iodine deposition because it predicts I<sub>2</sub> dry deposition depending on atmospheric turbulence parameters and surfaces properties of plant cover and may be used to learn about parametric influences. The model developed in this study will be tested on releases of gaseous radioactive iodine (<sup>129</sup>I) and under different conditions, particularly a longer release time (2 weeks) and a greater distance between the release point and the sampling site (2 km).

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### Appendix 1: Atmospheric iodine concentrations on filters, in blank NaOH and in NaOH samples during MIOSEC 2.

Date and time	Atmospheric iodine concentration (µg.L <sup>-1</sup> ) on filters in NaOH 0.5 N	Atmospheric iodine concentration (µg.L <sup>-1</sup> ) in blank NaOH 0.5 N	Atmospheric iodine concentration (µg.L <sup>-1</sup> ) in samples in NaOH 0.5 N	Volume (L) of NaOH 0.5 N in flask for atmospheric iodine samples	Atmospheric iodine concentration (µg)	Volume (L) of air sampled	Atmospheric iodine concentration (µg.m <sup>-3</sup> )
19/09/2018 10:32	< 2.5	< 2.5	66.3	0.1	6.6	60	110.6
19/09/2018 15:00	< 2.5	< 2.5	17.8	0.1	1.8	60	29.7
20/09/2018 09:30	< 2.5	< 2.5	11.0	0.1	1.1	60	18.4
20/09/2018 14:32	< 2.5	< 2.5	29.2	0.1	2.9	60	48.7
20/09/2018 17:30	< 2.5	< 2.5	18.5	0.1	1.9	60	30.9
21/09/2018 15:00	< 2.5	< 2.5	27.9	0.1	2.8	60	46.5
21/09/2018 18:30	< 2.5	< 2.5	70.0	0.1	7.0	60	116.7
22/09/2018 09:30	< 2.5	< 2.5	70.5	0.1	7.1	60	117.6
22/09/2018 17:00	< 2.5	< 2.5	9.1	0.1	0.9	60	15.2

24/09/2018 10:30	< 2.5	< 2.5	12.5	0.1	1.3	60	20.9
24/09/2018 16:30	< 2.5	< 2.5	9.6	0.1	1.0	60	16.1
25/09/2018 09:30	< 2.5	< 2.5	2.8	0.1	0.3	60	4.7
26/09/2018 10:00	< 2.5	< 2.5	3.0	0.1	0.3	60	4.9
27/09/2018 10:30	< 2.5	< 2.5	79.7	0.1	8.0	60	132.9

**Appendix 2:** Atmospheric iodine concentrations on filters, in blank NaOH and in NaOH samples during MIOSEC 3.

Date and time	Atmospheric iodine concentration ( $\mu\text{g.L}^{-1}$ ) on filters in NaOH 0.5 N	Atmospheric iodine concentration ( $\mu\text{g.L}^{-1}$ ) in blank NaOH 0.5 N	Atmospheric iodine concentration ( $\mu\text{g.L}^{-1}$ ) in samples in NaOH 0.5 N	Volume (L) of NaOH 0.5 N in flask for atmospheric iodine samples	Atmospheric iodine concentration ( $\mu\text{g}$ )	Volume (L) of air sampled	Atmospheric iodine concentration ( $\mu\text{g.m}^{-3}$ )
04/06/2019 16:05	< 2.5	< 2.5	7,1	0,1	0,7	60	11,8
04/06/2019 17:32	< 2.5	< 2.5	5,3	0,1	0,5	60	8,8
06/06/2019 07:35	< 2.5	< 2.5	26,7	0,1	2,7	60	44,6
06/06/2019 10:30	< 2.5	< 2.5	7,9	0,1	0,8	60	13,2
06/06/2019 12:30	< 2.5	< 2.5	11,3	0,1	1,1	60	18,9
06/06/2019 16:03	< 2.5	< 2.5	7,4	0,1	0,7	60	12,3
07/06/2019 09:55	< 2.5	< 2.5	16,9	0,1	1,7	60	28,2
07/06/2019 10:47	< 2.5	< 2.5	9,2	0,1	0,9	60	15,3

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