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The Role of Vegetation in Enhancing Radon Concentration and Ion Production in the Atmosphere.

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Abstract

The role of ions in the production of atmospheric particles has gained wide interest due to their profound impact on climate. Away from anthropogenic sources, molecules are ionized by alpha radiation from radon exhaled from the ground and cosmic gamma radiation from space. These molecular ions quickly form into ‘cluster ions’, typically smaller than about 1.5 nm. Using our measurements and the published literature, we present evidence to show that cluster ion concentrations in forest areas are consistently higher than outside. Owing to the low range of alpha particles, radon present deep in the ground cannot directly contribute to the measured cluster ion concentrations. We propose an additional mechanism whereby radon, which is water soluble, is brought up by trees and plants through the uptake of groundwater and released into the atmosphere by transpiration. We estimate that, in a forest comprising eucalyptus trees spaced 4 m apart, trees may account for up to 37% of the radon that is released from the ground during the middle of the day when transpiration rates are high. The corresponding percentage on an annual basis is 4.1%. Considering that 24% of the earth’s land area is still covered in forests; these findings have potentially important implications for atmospheric aerosol formation and climate.

Keywords: *Radon, cluster ions, atmospheric ions, environmental radioactivity, atmospheric aerosols*

1. Introduction

Radon-222 is a gaseous radioactive product of Radium-226 that is present in varying amounts in the lower atmosphere. It decays with a half life of 3.82 days into a series of short-lived progeny, which together, are the most significant contributors to human exposure from natural sources (1). Most of the radon is normally trapped in the ore-bearing rock deep within the earth but is also found closer to the surface where it migrates freely through rock faults and loose soil to enter the atmosphere. Radon has a relatively high solubility in water with a mole fraction solubility of 1.67×10^{-4} at 25ºC and 1 atm, which is higher than that of most gases found in normal air except carbon dioxide (2). Ground water in wells and springs often contain much higher concentrations of radon than surface water in rivers and lakes. Typically, surface water contains about 0.4 Bq L^{-1} while ground water has about 20 Bq L^{-1} (3). The average radon level in ground water in the US is 13 Bq L^{-1} (4). These levels are determined by several factors including rock type and can vary by up to two orders of magnitude. Levels as high as 370 Bq L^{-1} have been detected in ground water moving through granite while, at the other extreme, they can be as low as 3 Bq L^{-1} in the presence of sedimentary rocks and in coastal plains (5).

One square metre of typical soil containing 30 Bq kg^{-1} of radium will release between 1000 and 2000 Bq of radon to the atmosphere each day, with a global average of about 15 mBq m^{-2} s^{-1} (6,7). Assuming an atmospheric half-depth of 700 m, an average exhalation of 20 mBq m^{-2} s^{-1} gives a ground level radon concentration of 8 Bq m^{-3} (8). This is in good agreement with ambient values of 5-20 Bq m^{-3} found in various locations around the world (6).

In the natural environment, cluster ions are formed mainly by cosmic radiation, gamma radiation from the ground and alpha radiation from radon and its short-lived decay products in the air. Thus, we expect the cluster ion concentration to be directly related to the radon level. This is wellillustrated by measurements within an underground cavern with high radon exhalation in the Czech Republic (9) that showed a strong correlation between the radon levels and the positive ions (R^2=0.89) and the negative ions (R^2=0.96). In a previous study (10), we showed that the median cluster ion concentration in woodland areas (725 cm^{-3}) was significantly higher than in open parks (269 cm^{-3}). The relatively high values found in woodland areas that are reasonably far from human activities are in agreement with exceptionally high values that have been found at remote forest locations. For example, values over 1000 cm^{-3} have been regularly reported from the boreal forests in Northern Europe (11-14). Suni et al. (15) measured average cluster ion concentrations of over 4000 cm^{-3} in a Eucalyptus forest in Tumbarumba, NSW, Australia, over a period of 16 months. They suggested that these exceptionally high concentrations were a result of strong radon efflux from the soils around the field site but were unable to confirm this. There is very little information on radon concentrations in forest air. Suni et al. (15) and Martens et al. (16) reported levels of 9-102 Bq m^{-3} and 13-24 Bq m^{-3} in Tumbarumba and the Amazon forest, respectively. However, Hirsikko et al. (17) did not find levels greater than 11 Bq m^{-3} in a boreal forest in Finland.

In this paper, we present evidence for an alternative explanation for the occurrence of high radon and cluster ion concentrations in forested areas based on a secondary pathway of radon efflux from the ground through the uptake and transpiration of ground water by vegetation.

2. Methods

The aim of this study was to support our hypothesis by (a) gathering and analysing the evidence available in the literature and (b) carrying out some new measurements, as described below.

Literature evidence: An extensive review of the literature was carried out to investigate the present state of knowledge on the following aspects and questions: what are the typical cluster ion concentrations in forest environments and how do they compare with that in other locations away from human activities, what are the typical radon exhalation rates from the ground in forest areas and are they different to other areas, and what are the typical radon concentrations in the atmosphere, in soils and in ground water? We collated data from 15 studies conducted in forest locations or non-forested open areas, away from busy roads and industries, over the past 10 years that reported both positive and negative cluster ion concentrations. We also collated data from 18 papers that have reported rates of radon exhalation from the ground. Studies carried out at sites showing unnaturally high exhalation rates due to the presence of uranium mill tailings in the ground were avoided. Six of the 18 studies were in forest locations. Having established these findings, we set out to investigate how cluster ion concentrations are related to environmental radon concentrations. Next, we considered the typical water uptake and transpiration rates of trees and plants to estimate how much radon is exhaled from vegetation.

Experimental study: In order to support the evidence in the literature, we carried out measurements of cluster ions, over a total time period of about 150 h, inside and outside several forest locations as well as at other locations away from human activities. Three large forest areas in South East Queensland, Australia, were selected. These were (a) Mount Coot-tha Forest Reserve - over 1,500 ha of open eucalypt forest in the Taylor Range which forms a backdrop of hills to the city of Brisbane (b) Daisy Hill State Forest - situated south of Brisbane covering about 430 ha and (c) Brisbane Forest Park – a large 26 500 ha area. The vegetation in all three areas is mainly associated with dry eucalypt forest including Spotted gum (Corymbia varigata), Grey gum (Eucalyptus propinqua), Forest red gum (Eucalyptus tereticornis) and Narrow-leaved ironbark (Eucalyptus crebra). Native grasses, primarily Kangaroo grass (Themeda triandra) and Blady grass (Imperata cylindrica) make up the minimal ground cover.

Measurements outside the forests were carried out on the upwind sides of the forests. All measurements were restricted to daylight hours to avoid the marked diurnal variations in atmospheric ion concentrations (18). Sampling was carried out at a height of 1 m above the ground under fair weather conditions, with the air temperature between 20ºC to 30ºC. Cluster ion concentrations were measured with Alphalab air ion counters. This instrument has a dynamic range of 10 – 10^6 ions cm^{-3} with a minimum detectable charge concentration of 10 ions cm^{-3} and a response time of 2 s at a volume sampling rate of 0.8 L s^{-1}. The minimum characterisable mobility of the unit is 0.5 cm^2 V^{-1} s^{-1}, which corresponds to a detectable maximum ion size of 1.6 nm. The instrument has the capability of monitoring negative and positive ions separately, but not simultaneously. Hence, two instruments were used to measure both positive and negative ions separately at each measurement point. The instruments were interfaced to a laptop computer that logged the data in real time at 1 s intervals.

Significant differences between population means were assessed using a Students t-test at a confidence level of 95%.

3. Results and Discussion

Cluster ion concentrations: Our measurements showed that, at any given time of day, the mean cluster ion concentration inside a forest was greater than the corresponding value immediately outside on its upwind side. Fig 1 shows a short time series of the total cluster ion concentrations measured on a relatively calm, sunny day, inside the Daisy Hill Forest Park and at its upwind edge. The wind speed was 1-2 m s^{-1}. The difference between the outside and the inside of the forest in this case was at least a factor of 2. The average value of all our measurements in the forests ranged from 700 cm^{-3} to 2000 cm^{-3} with a mean of 1600 cm^{-3} which was significantly higher than the corresponding value of 670 cm^{-3} found upwind of all the forested sites.

Literature data: Our analysis of the literature data (Fig 2) showed that the mean cluster ion concentration reported from the forest sites (1649 ± 1071 cm^{-3}) was three times greater than the corresponding value for the open sites (494 ± 188 cm^{-3}), where the uncertainties indicate the respective standard deviations. A Students t-test showed that the two means were significantly different at a confidence level of 95%. Included in this figure are our measurements of cluster ion concentrations inside and outside forest areas, labeled 'Present Study'.

Following these observations, it was instructive to see if radon concentrations in forest areas, reported in the literature, were greater than outside. Unfortunately, we could find very little information on radon concentrations in forest air other than for three studies (15,16,17), which all showed values greater than the global average. Considering this limitation, we looked at radon exhalation rates from the ground within and outside forests (Fig 3). The Wilkening et al. (7) and Schery et al (19) values represent worldwide and Australian averages of 15 and 22 mBq m^{-2} s^{-1}, respectively. The mean values for the open and forest sites were 17.8 ± 7.0 mBq m^{-2} s^{-1} and 15.6 ± 10.9 mBq m^{-2} s^{-1}, respectively. The conclusion from our analysis was that, unlike for the ion concentrations, the t-test showed no significant difference between the mean radon exhalation rates from the ground for the open and forest sites.

The hypothesis: The statistically significant difference in cluster ion concentrations between forest measurement sites and other sites (Fig 2), coupled with the strong correlation between cluster ion concentration and radon levels in the atmosphere (9) and the absence of any difference in radon exhalation from the ground between the two site types (Fig 3) suggest the existence of another source for the radon present in forest air. This source cannot be the external gamma radiation from the ground or cosmic radiation as these two factors are not expected to be different at the two types of locations. A likely source is the trees and vegetation that form the forests. Thus, we propose a new hypothesis based on the release of radon into the atmosphere through the uptake and transpiration of groundwater by trees. In well-wooded areas, transpiration from trees can account for over 80% of total evapotranspiration (20). In dry climates, a large fraction of the water transpired by trees comes from deep within the ground. For example, native Australian trees, such as eucalyptus, have deep root systems that can extend down to 15 m, often reaching the water table, and can transpire nearly 200 lit of water a day (21). A study of a Eucalyptus Camaldulensis in Australia (22) showed that the average annual water uptake exceeded the rainfall by a factor of 2.7. As solar radiation and temperature play important roles, the rate of transpiration is a maximum in the middle of the day.

Radon in groundwater and uptake by vegetation: Groundwater is rich in radon. Uptake of groundwater by trees and release to the atmosphere by transpiration offers an alternative path to direct exhalation of radon from the ground with evidence in the literature for this process. Exhalation rates of Rn-222 from leaves of field corn have been found to exceed the typical exhalation rates from the ground by a factor of about six (23). Martell (24) has cited this study in suggesting that, during periods with high rates of plant transpiration, the flux of Rn-222 is some 5-10 times the average flux for bare soils. Lewis and MacDonell (25) measured exhalation rates from 8 to 28 mBq s^{-1} m^{-2} of leaf area from corn, sunflower and tall fescue grown in uranium mill tailings solids and suggested that radon was transported from the rooting medium to leaves by mass flow in liquid water. The Rn-222/Ra-226 ratio in the above-ground plant organs can be 25 to 185-fold higher as compared to that of the root-containing soil layer (26) and may be attributed to the preferential uptake of radon by plants. Radon in the ground is influenced by vegetation with concentrations at a depth of 0.5m in bare plots being significantly higher than in vegetated plots, while the removal of radon through the uptake of groundwater can result in a depletion of radon concentration in the ground near a tree (27) (Fig 4). The figure shows the Rn-222 concentration in the ground at a depth of 0.8 m as a function of distance from a large oak tree that is (a) leafless and (b) coming into leaf. In both cases the Rn-222 concentration decreased on approaching the tree. On the global scale, the mean Rn-222 flux from soil (15 mBq m^{-2} s^{-1}) is significantly smaller than the mean global removal rate of the long lived daughter Pb-210 from the atmosphere (25 mBq m^{-2} s^{-1}) (28). This imbalance may be attributed to the direct release of radon from vegetation.

Radon exhalation calculation: In order to support our hypothesis, we performed a simple calculation based on the transpiration rate of a typical eucalyptus tree during the middle of the day. Considering that such a tree can transpire about 100-200 litres of water per day (21), we assumed a peak transpiration rate of 20 lit h^{-1} near mid-day. Assuming a ground water radon concentration of 20 Bq lit^{-1}, this gives a radon exhalation rate of approximately 111 mBq s^{-1}. If the trees were 4m apart, the ground area under each tree may be assumed to be a circle of radius 2 m giving an area of approximately 12.5 m^2. Assuming a radon exhalation rate of 15 mBq m^{-2} s^{-1}, the total exhalation from the ground under the tree is 187 mBq s^{-1}. This simple calculation suggests that, during the middle of the day when transpiration rates are at a maximum, although most of the Rn-222 in the atmosphere is still exhaled from the ground, as much as 37% may be exhaled by trees. This factor will be lower at other times of the day and in dry areas with low transpiration rates. Averaged over large areas, long-term evapotranspiration rates cannot exceed the rainfall rate. Therefore, taking the annual rainfall in Brisbane to be 1000 mm, the maximum volume of water that the tree can transpire in one year is 1.25 x 10^4 lit. Repeating the calculation on an annual basis the percentage of radon that is released by the tree is 4.1%.

At this point, it is pertinent to refer to a previously published study which plays down the importance of transpiration as an important source of radon transport to the atmosphere (29). In this study, the authors calculated the amount of radon that may be released by vegetation during transpiration by assuming that 25% of the radon present in soil pores will dissolve in rainwater seeping into the ground. Taking the soil gas Rn-222 concentration to be 1 pCi cm^{-3}, the concentration of Rn-222 in transpired water is 0.25 pCi cm^{-3} or 9.25 Bq lit^{-1}. For a climate with an annual rainfall of 100 cm they calculated that the amount of radon that is released by vegetation during transpiration was 8 x 10^{-4} pCi m^{-2} s^{-1}. This value is much smaller than the typical surface flux density of 1 pCi m^{-2} s^{-1} and, hence, they concluded that vegetation played a negligible role in releasing radon into the atmosphere. However, we would like to draw attention to three details in this calculation: (i) we believe that there is a numerical error in the calculated value of 8 x 10^{-4} pCi m^{-2} s^{-1} which should be 8 x 10^{-3} pCi m^{-2} s^{-1}, (ii) the concentration of Rn-222 in the transpired water (9 Bq lit^{-1}) is too low and (iii) the typical surface flux density of 1 pCi m^{-2} s^{-1} (37 mBq m^{-2} s^{-1}) is too high. Replacing the values of these two quantities by 20 Bq lit^{-1} and 15 mBq m^{-2} s^{-1}, respectively, yields a better estimate of 4.7% for the radon released by transpiration as a percentage of the total radon exhaled.

Neither the peak nor the annual enhancement is sufficient to explain the three-fold increase in cluster ion concentrations found when going from open areas into forests (Fig 2), indicating that there are other important factors. Soil porosity and moisture content are known to affect the radon exhalation rate from the ground (23,27,30). However, the null result in Fig 3 suggests that the effects of these factors on the exhalation rates of radon from the ground in forests may balance out. A more likely explanation is the role of trees in shielding the forest from winds that serve to dissipate the radon exhaled from the ground.

Diurnal variations: Radon is a gas that is heavier than air and in stable atmospheric conditions, such as that which occur during the night and in the early morning hours, it tends to accumulate close to the ground. Turbulence due to solar heating during the day cause large turbulent eddy diffusion in the atmosphere that can dilute radon emanating from the ground (8). Moreover, at a local level, there is good evidence to suggest that the radon concentration in the atmosphere follows a diurnal trend, similar to the cluster ion concentration, with a maximum in the early morning and a minimum in the afternoon. Fig 5 shows the diurnal variations of the radon levels derived from observations in

India (31) and in the Amazon (16). Both curves show the expected peak radon concentration in the morning. However, of more importance to our argument is the second peak that is observed in the afternoon. Two other studies have also reported this same phenomenon (30,32).

Since cosmic radiation does not show a significant diurnal variation (17,33), we expect the ion concentrations in the atmosphere to be directly related to the radon level. Our measurements of cluster ion concentrations near the Brisbane Forest Park showed a diurnal trend similar to that of radon with a maximum in the early hours of the morning, a minimum during the middle of the day, followed by a secondary maximum in the afternoon (Fig 6). Note maxima at 6 am and minima at 1 pm. This observation is in agreement with three other previous studies of the diurnal variation of cluster ions in the environment (17,34,35). Another study in a boreal forest has shown that the ion production rate, estimated from the radon concentration and external radiation, was a minimum close to noon and increased thereafter during the afternoon (33). Air temperature and air turbulence generally peaks during mid-afternoon and therefore both the radon concentration and the ion concentration are not expected to increase at this time (8). Our hypothesis offers an explanation based on the release of radon into the atmosphere by plant transpiration which generally peaks during the afternoon.

Global implications: Despite rapid deforestation, 24% of the earth’s land area is still covered by forests. Therefore, the findings of this study suggest that the distribution of ions over the earth’s surface may not be uniform. Ion-induced nucleation has been proposed as a possible mechanism for particle formation in the atmosphere (36). Aerosol particles potentially affect cloud formation and radiative transfer and play an important role in climate (37,38). While, it is known that vegetation is an important source of atmospheric particles through homogeneous nucleation of biogenic precursors (13,39), a possible role through ion-induced nucleation has not been investigated in-depth. There is a positive relationship between radon concentration and ion production in the atmosphere (9,11,33,35). We have shown that a significant enhancement of radon release from vegetation can occur during times of peak transpiration. This usually takes place near mid-day, which is also the time when aerosol production by nucleation is at its maximum (33,39). Therefore, vegetation cover and its distribution around the globe may be important in other unexpected ways in atmospheric aerosol formation and climate change, and should be taken into account in atmospheric particle production models.

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Figure Legends

Fig 1: Short time series of cluster ion concentrations measured on a calm, sunny day, inside a forest park (upper curve) and outside the forest park on its upwind edge (lower curve).

Fig 2: Average positive and negative cluster ion concentrations in open areas and forest areas from the present study, shown alongside all other results as reported in the recent literature.

Fig 3: Radon exhalation rates from the ground at open and forest sites as reported in the literature with mean values which, unlike for the ion concentrations, were not significantly different.

Fig 4: Radon-222 concentration in the ground as a function of distance from a large oak tree that is (a) leafless and (b) coming into leaf. Data extracted from Feige and Wiegand (27).

Fig 5: Diurnal variation of the radon level in the air plotted from data reported by Martens et al. (14) and Chandrashekara et al. (31).

Fig 6: Typical diurnal variation of the positive, negative and total cluster ion concentrations monitored on a clear, calm day, in a wooded area bordering the Brisbane Forest Park in August 2010.

Colour Figures for On-Line Edition

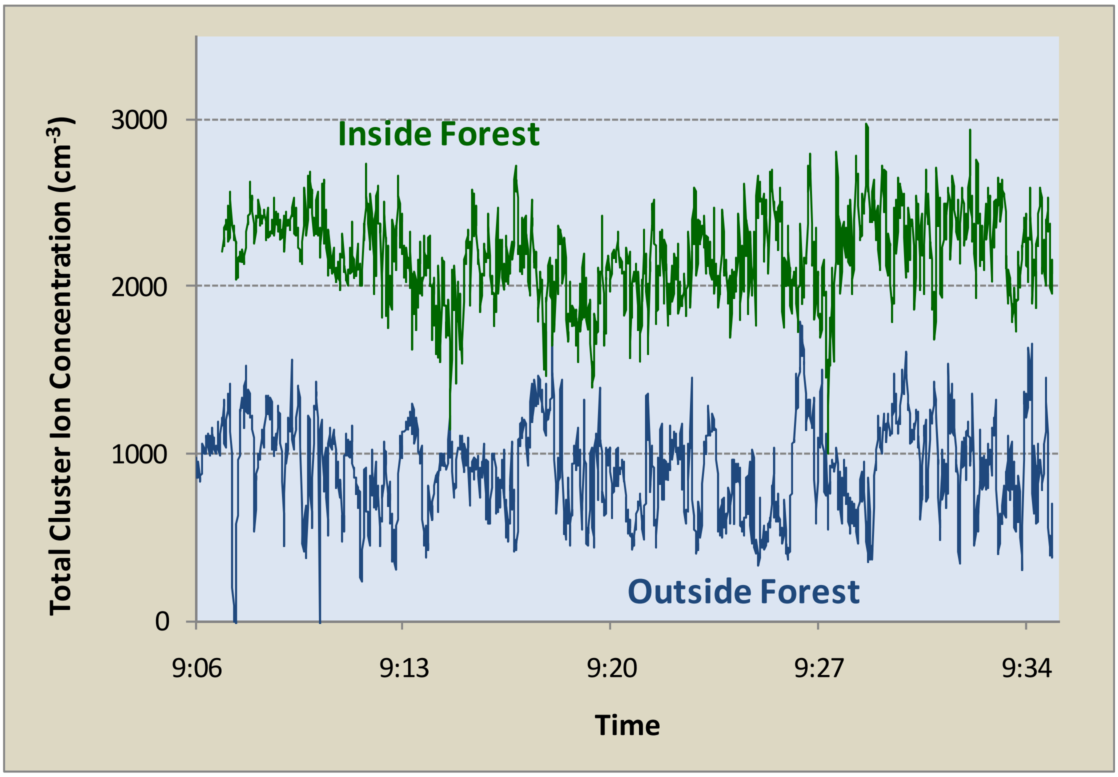


Fig 1

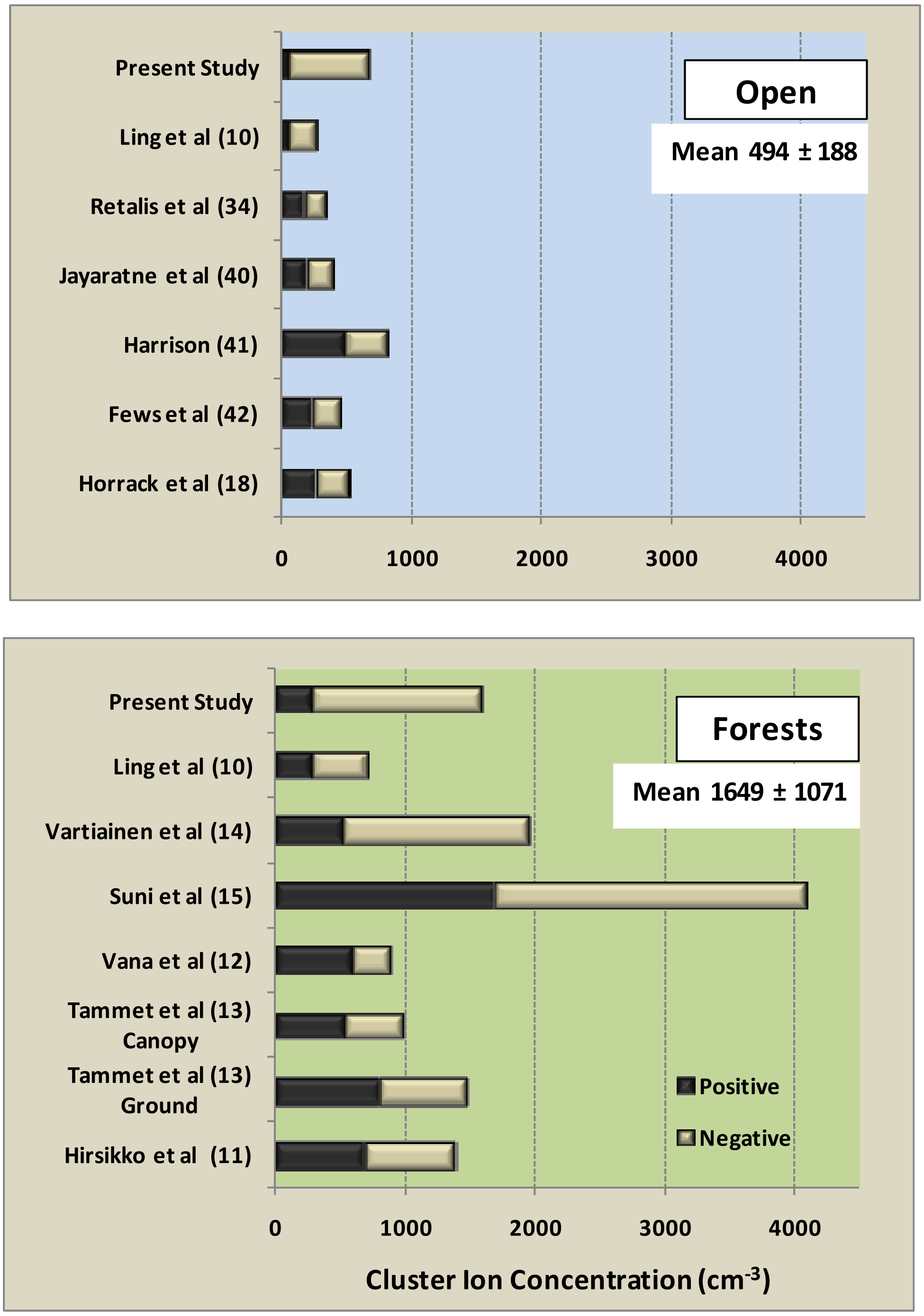

Open
Mean 494 ± 188
Present Study
Ling et al (10)
Retalis et al (34)
Jayaratne et al (40)
Harrison (41)
Fews et al (42)
Horrack et al (18)
0
1000
2000
3000
4000
Forests
Mean 1649 ± 1071
Present Study
Ling et al (10)
Vartiainen et al (14)
Suni et al (15)
Vana et al (12)
Tammet et al (13) Canopy
Tammet et al (13) Ground
Hirsikko et al (11)
■ Positive
▨ Negative
0
1000
2000
3000
4000
Cluster Ion Concentration (cm⁻³)

Fig 2

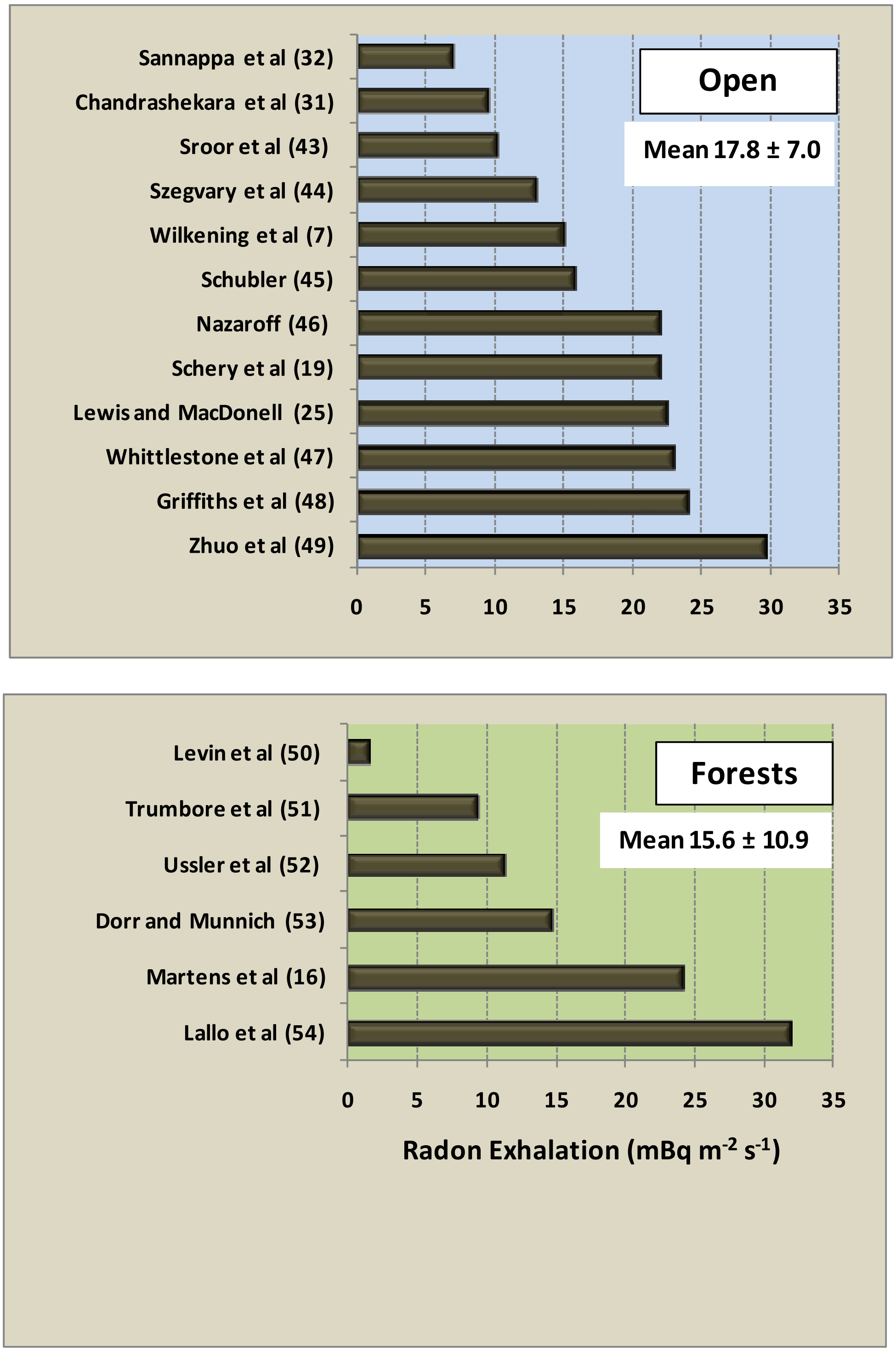

Sannappa et al (32)
Chandrashekara et al (31)
Sroor et al (43)
Szegvary et al (44)
Wilkening et al (7)
Schubler (45)
Nazaroff (46)
Schery et al (19)
Lewis and MacDonell (25)
Whittlestone et al (47)
Griffiths et al (48)
Zhuo et al (49)
Open
Mean 17.8 ± 7.0
0
5
10
15
20
25
30
35
Levin et al (50)
Trumbore et al (51)
Ussler et al (52)
Dorr and Munnich (53)
Martens et al (16)
Lallo et al (54)
Forests
Mean 15.6 ± 10.9
0
5
10
15
20
25
30
35
Radon Exhalation (mBq m-2 s-1)

Fig 3

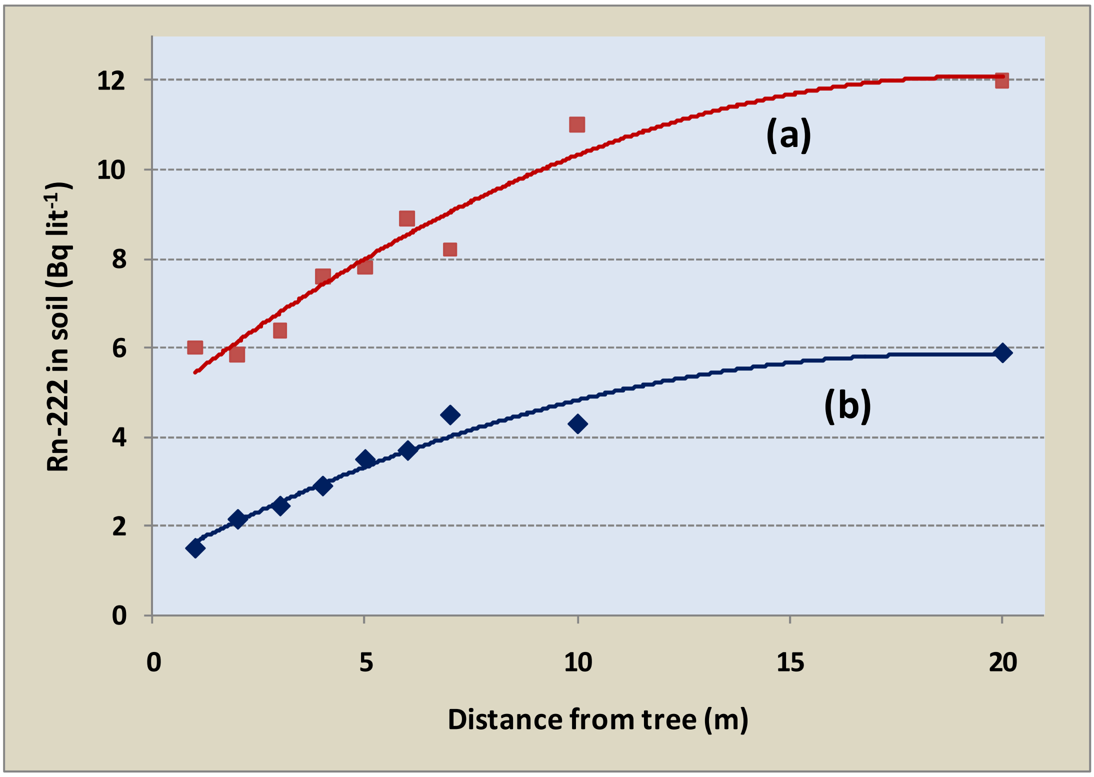

Rn-222 in soil (Bq lit^{-1})
Distance from tree (m)
0
2
4
6
8
10
12
0
5
10
15
20
(a)
(b)

Fig 4

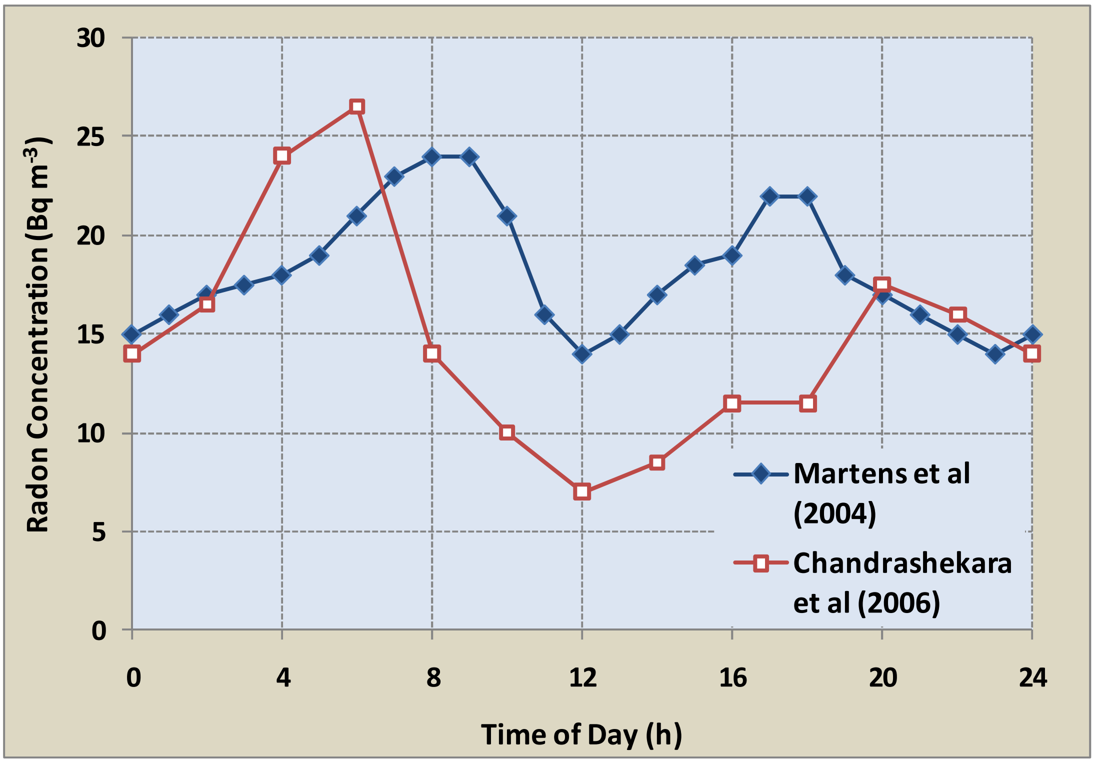

30
25
20
15
10
5
0
Radon Concentration (Bq m^{-3})
0
4
8
12
16
20
24
Time of Day (h)
Martens et al (2004)
Chandrashekara et al (2006)

Fig 5

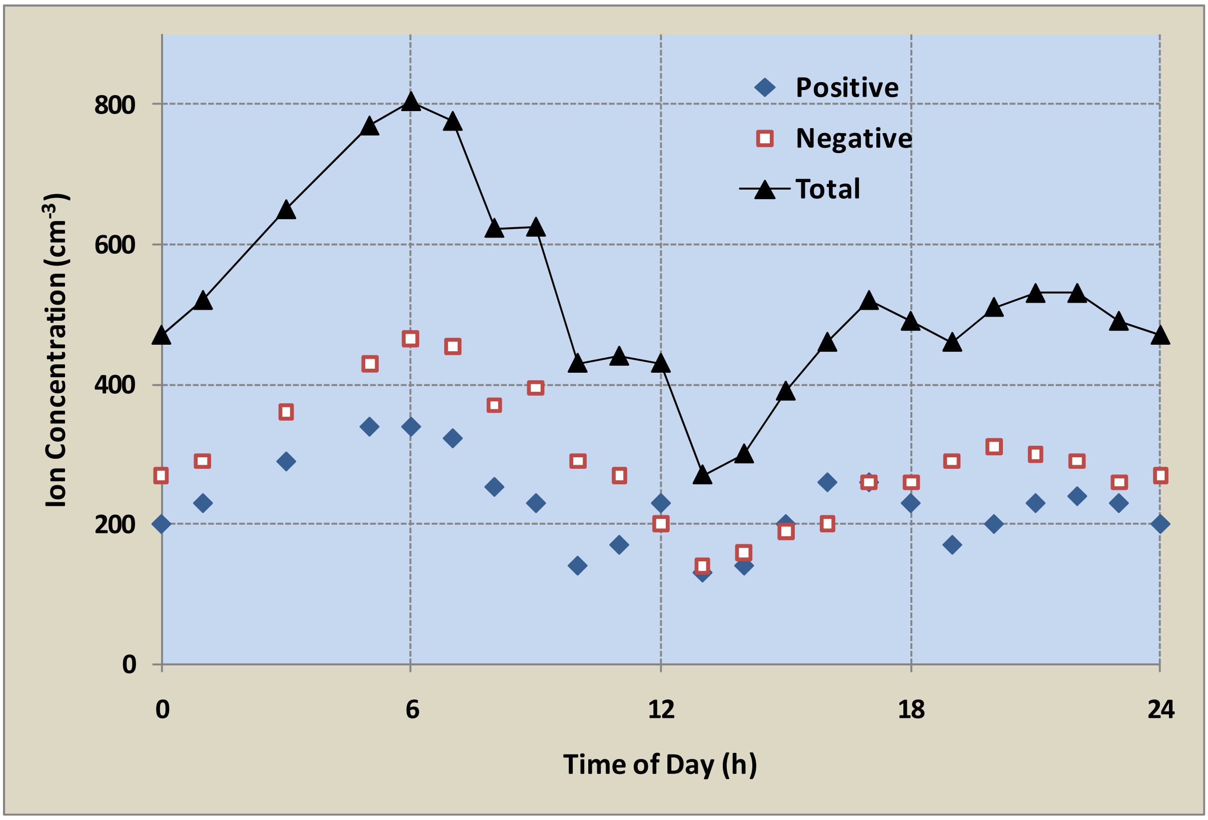

Positive
Negative
Total
Ion Concentration (cm^{-3})
0
200
400
600
800
0
6
12
18
24
Time of Day (h)

Fig 6

Black and White Figures for Print Edition

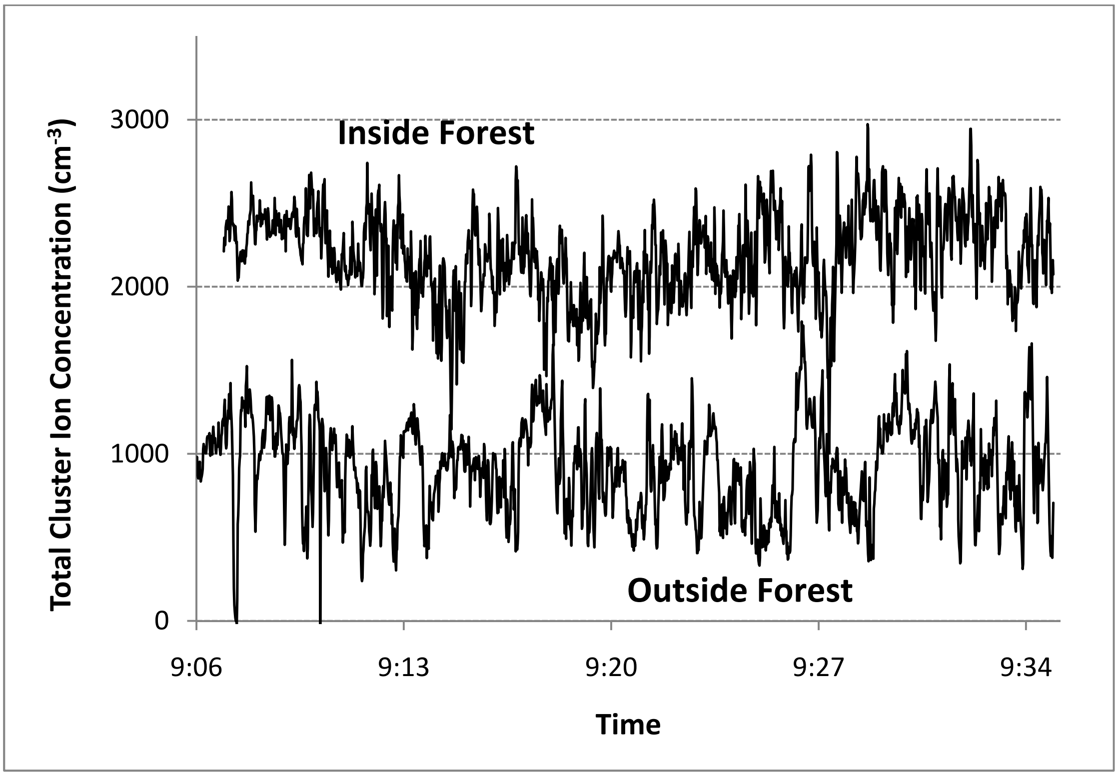
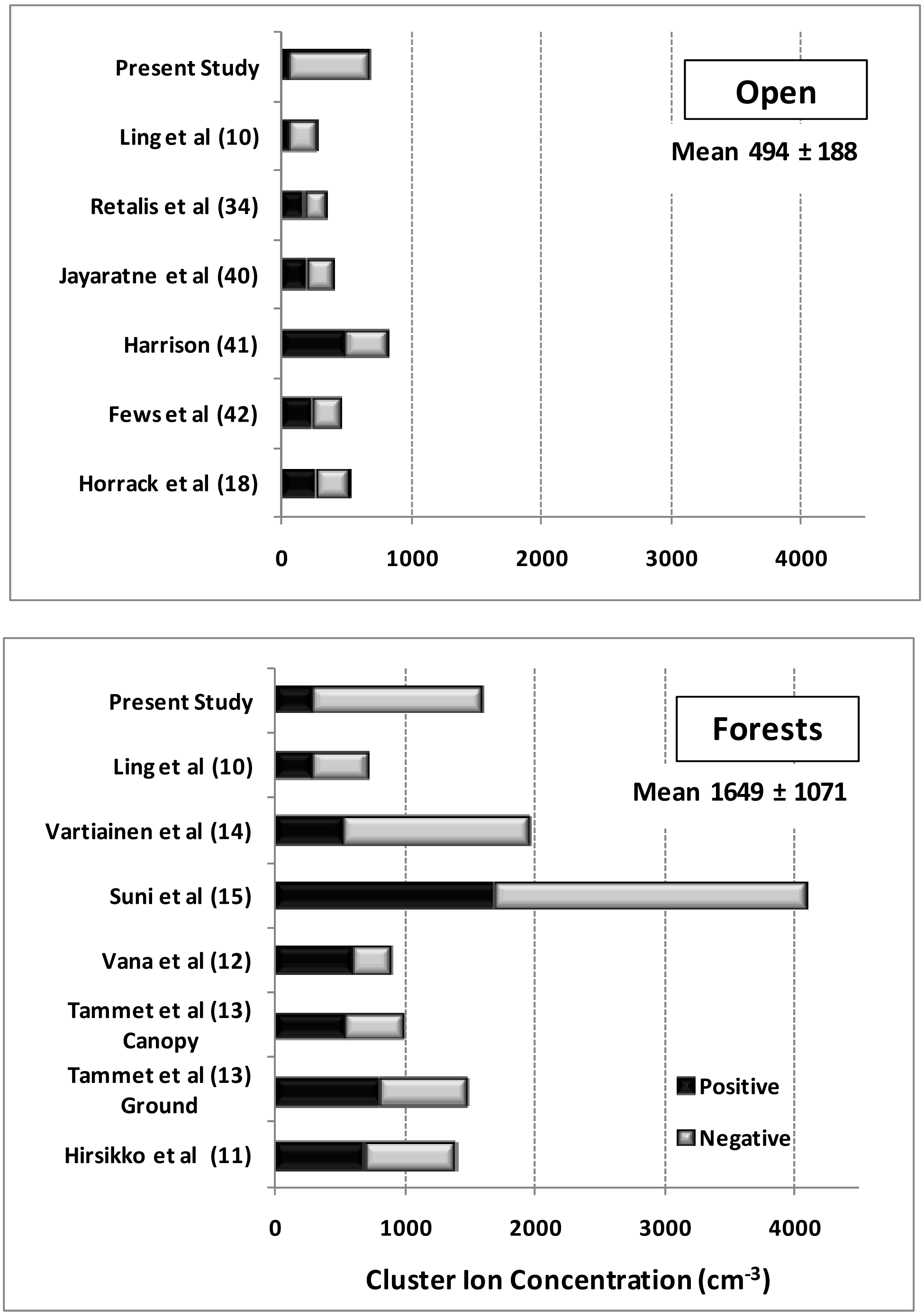


Fig 1


Present Study
Ling et al (10)
Retalis et al (34)
Jayaratne et al (40)
Harrison (41)
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Horrack et al (18)
0
1000
2000
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Open
Mean 494 ± 188
Present Study
Ling et al (10)
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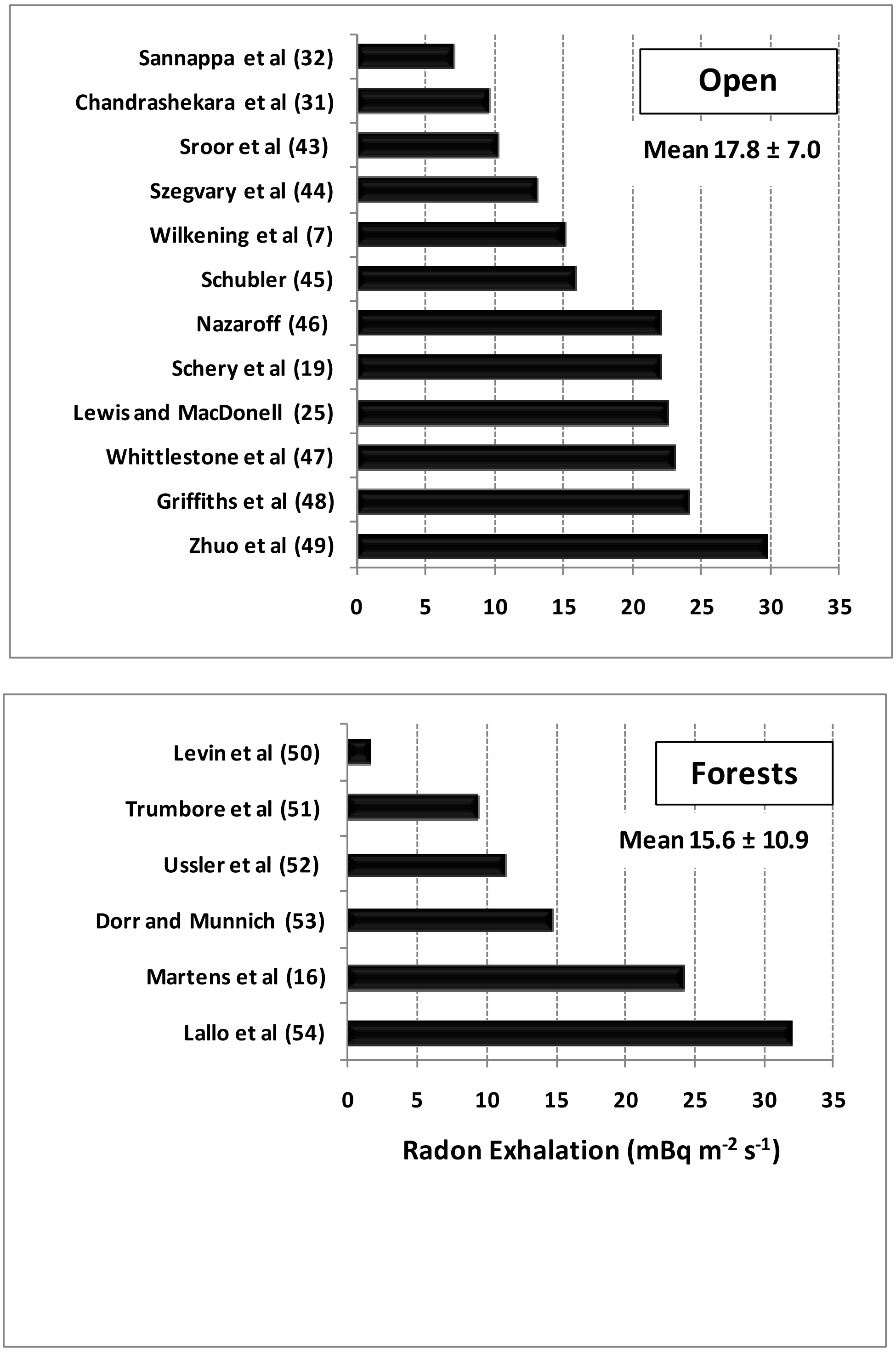
Suni et al (15)
Vana et al (12)
Tammet et al (13) Canopy
Tammet et al (13) Ground
Hirsikko et al (11)
0
1000
2000
3000
4000
Forests
Mean 1649 ± 1071
■ Positive
▨ Negative
Cluster Ion Concentration (cm^{-3})

Fig 2


Open
Mean 17.8 ± 7.0
Sannappa et al (32)
Chandrashekara et al (31)
Sroor et al (43)
Szegvary et al (44)
Wilkening et al (7)
Schubler (45)
Nazaroff (46)
Schery et al (19)
Lewis and MacDonell (25)
Whittlestone et al (47)
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0
5
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Forests
Mean 15.6 ± 10.9
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Dorr and Munnich (53)
Martens et al (16)
Lallo et al (54)
0
5
10
15
20
25
30
35
Radon Exhalation (mBq m-2 s-1)

Fig 3

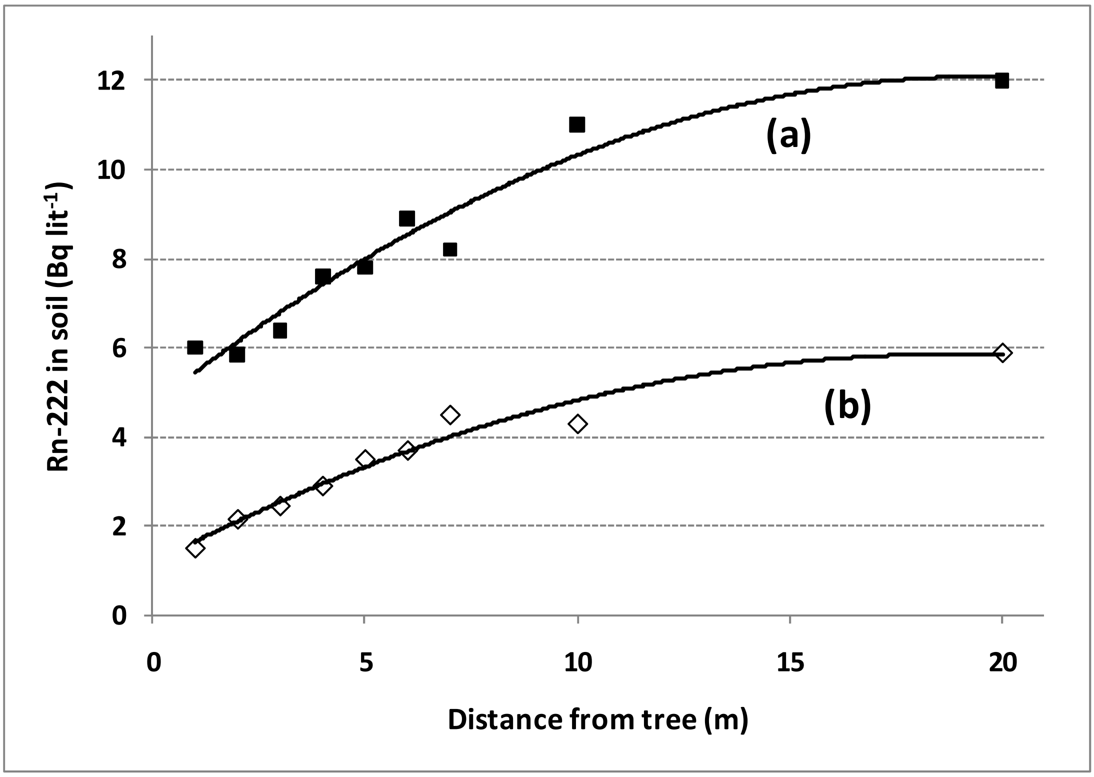
12
10
8
6
4
2
0
Rn-222 in soil (Bq lit^{-1})
0
5
10
15
20
Distance from tree (m)
(a)
(b)

Fig 4

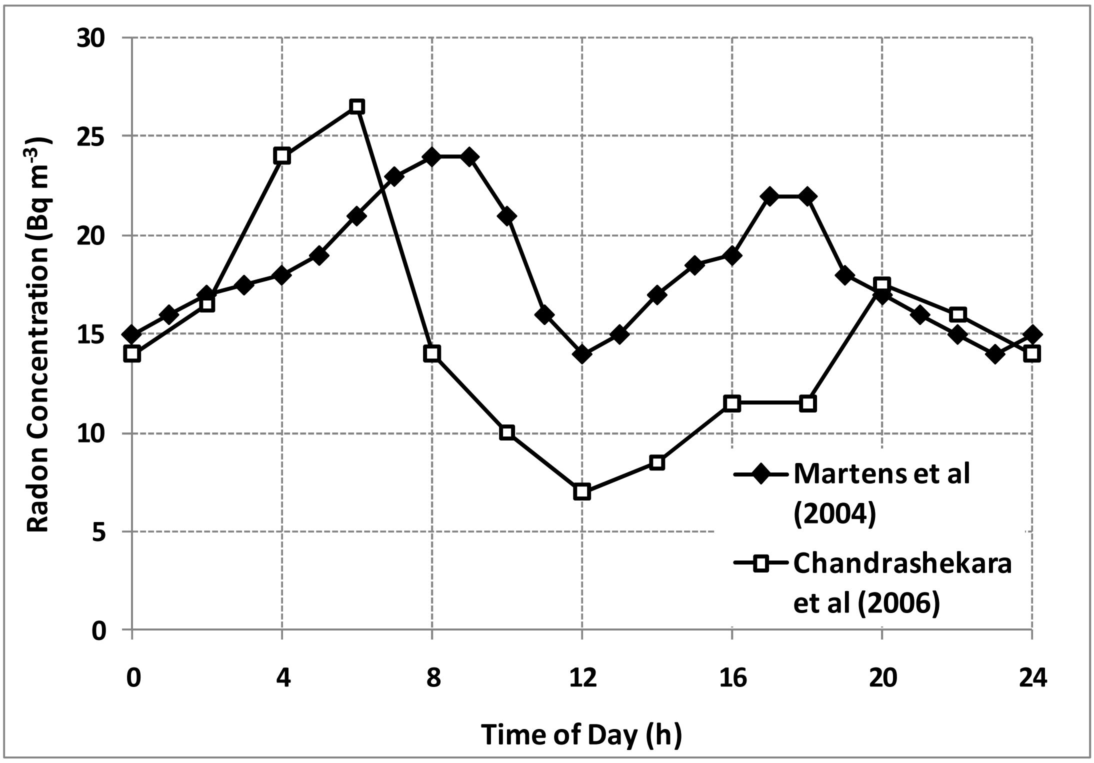
Radon Concentration (Bq m⁻³)
30
25
20
15
10
5
0
0
4
8
12
16
20
24
Time of Day (h)
Martens et al (2004)
Chandrashekara et al (2006)

Fig 5

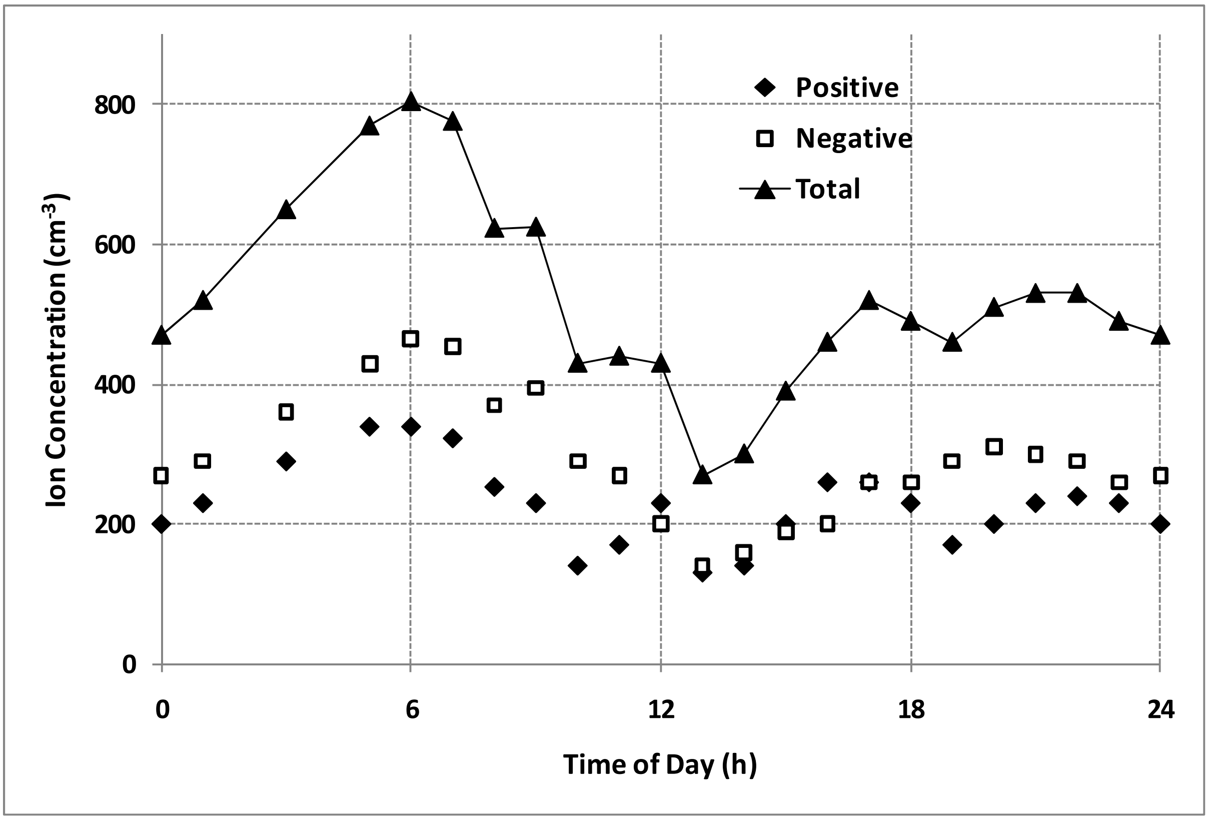
Positive
Negative
Total
Ion Concentration (cm-3)
800
600
400
200
0
0
6
12
18
24
Time of Day (h)

Fig 6