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学位の種類	博　士（理　学）
学位記番号	理博第１８３７号
学位授与年月日	平成１３年３月26日
学位授与の要件	学位規則第４条第１項該当
研究科，専攻	東北大学大学院理学研究科（博士課程）地球物理学専攻
学位論文題目	Variations of atmospheric components over the past 340000 years from Dome Fuji deep ice core,Antarctica （南極ドームふじ深層氷床コアから推定された過去34万年にわたる大気組成の変動）
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Abstract

A 2500 m-deep ice core drilled at Dome Fuji, Antarctica was analyzed for concentrations of CH_4, N_2O and CO_2, $\delta^{15}N$ of N_2, $\delta^{18}O$ of O_2 and $\delta(O_2/N_2)$ in the atmosphere, as well as total air content in ice, by using a wet extraction method. The analytical precision for the respective species were estimated to be ±6 ppbv, ±3 ppbv, ±1 ppmv, ±0.02 ‰, ±0.04 ‰, ±0.2 ‰ and ±0.6 mlSTP kg^{-1}. Air samples were also collected from firn at Dome Fuji and H72, Antarctica and then analyzed for the same species as above, to allow an accurate estimation of the bubble close-off depth. For dating the air occluded in the Dome Fuji ice core, the age difference between ice and air (Δ age) were estimated by model calculation. Based on these analytical and model studies, precise concentration variations of CH_4, N_2O and CO_2, $\delta^{15}N$ of N_2, $\delta^{18}O$ of O_2 and $\delta(O_2/N_2)$ with a time resolution of about 1 kyr over the past 340 kyrs covering three glacial-interglacial cycles were deduced. Dole effect, which is a proxy of terrestrial and marine production, was also deduced from atmospheric $\delta^{18}O$ and $\delta^{18}O$ of seawater from the deep sea core. The results obtained in this study are summarized as follows:

1) The molecular diffusion was the dominant mechanism for gas movement in firn, resulting in gravitational separation and age delay of gas components. In addition to this effect, non-diffusive transport of gas was also important at H72, due to sealing of the dense winter layer near the top of the bubble close-off zone. The convective mixing zone was found to be 9 and 3 m at Dome Fuji and H72, respectively. Using the diffusion model, Δ age was found to be 2200 years at Dome Fuji and 140 years at H72 under the present condition. The width of air age in ice was estimated to be 500 years at most at Dome Fuji and 20 years at H72.

2) The close-off depth at Dome Fuji was fairly stable in the Holocene epoch, with an average of 100.3±0.5 m, and deeper by 15 m in the glacial period. Δ age was highly variable over the glacial-interglacial cycles, ranging between 2000 and 5000 years, with high values in the glacial maximum. Such a large fluctuation was ascribed primarily to the changing accumulation rate and secondarily to the changing close-off depth.

3) $\delta^{15}N$ of air in the Dome Fuji core was high in the interglacial period, with a mean value of 0.51 ‰, and decreased to 0.45 ‰ in the glacial maximum, which is opposite to that expected from the densification theory. The close-off depth at the Last Glacial Maximum was estimated to be 78 m on the basis of the measured $\delta^{15}N$ values,

while the densification model calculated it 116 m. This fact suggests that the densification model possibly overestimated Δ age by 1.4 kyrs in the glacial maximum and less in the rest of the period.

4) The CH_4 concentration increased rapidly from 360-420 to 700-730 ppbv during the glacial-interglacial transitions. After the interglacial period, the concentration decreased rapidly to 470-490 ppbv and then gradually toward the lowest value in the glacial maximum, showing a number of fluctuations with amplitudes of more than 100 ppbv. The CH_4 concentration variation was found to be concomitant with the temperature variation, which suggests that natural CH_4 sources especially in the region from the tropics to northern latitudes were affected by climate change. The CH_4 concentrations from the Dome Fuji core showed similar temporal variations to those from other Antarctic ice cores, with some systematic difference of the concentration. However, detailed comparison with the Vostok record revealed somewhat large differences in some periods of the last 340 kyrs. It was also found that almost all prominent concentration peaks seen in the Dome Fuji CH_4 record of the last glacial period were identified in the Taylor Dome record. Based on the measured CH_4 concentrations, CH_4 source was found to be stronger in the glacial maximum at around 255 kyrs BP by 13 % than in other glacial maxima. It was also found that CH_4 emission sometimes occurred in the region from the tropics to northern high latitudes, being independent of climate change in southern high latitudes.

5) The N_2O concentration showed clear evidence for glacial-interglacial variations, with the respective concentrations 260-275 and 230-240 ppbv for the interglacial and glacial periods. The N_2O concentration also occasionally showed considerably high values in the glacial maximum. Temporary rapid increases of the N_2O concentration were synchronized with those of the CH_4 concentrations, which suggests that N_2O sources in the region from the tropics to northern high latitudes were activated in those times. On the other hand, clear negative correlation of the CH_4 concentration with the N_2O concentration was found in the middle of the Holocene, due probably to enhanced oceanic N_2O sources. During the Younger Dryas period, the N_2O concentration was lowered by 35 ppbv, in response to climate change in northern high latitudes.

6) The CO_2 concentration varied between 190 and 300 ppmv showing clear glacial-interglacial variations. Spectral analysis of the time series of the CO_2 concentration showed that very strong periodicity with 115 kyrs cycle^{-1} is predominant. From the comparison with the CO_2 concentration data from other ice core studies for the past 50 kyrs, it was found that our value obtained using the wet extraction method reconstructed the atmospheric history of CO_2 fairly well. The CO_2 concentration variations deduced from the Dome Fuji core for the past 340 kyrs were similar to those from the Vostok core as a whole.

7) The atmospheric $\delta^{18}O$ record deduced from the Dome Fuji core and the SPECMAP record for $\delta^{18}O$ of seawater were correlated fairly well with each other during 0-220 kyrs BP. However, the $\delta^{18}O$ record of the Dome Fuji core preceded SPECMAP record before 220 kyrs BP. By comparing our $\delta^{18}O$ record with that from the Vostok core, it was also found both records are in good agreement with each other, except for phase in some periods. From the variation of Dole effect deduced for the penultimate interglacial period, it was suggested that primary production of the terrestrial biosphere was enhanced especially during 123-114 kyrs BP.

8) The total air content in the Dome Fuji core varied between 80 and 95 ml kg^{-1}, showing rapid decrease at terminating of all glacial periods. The past pressure values at the bubble close-off depth (P_c), calculated from the total air content and the temperature at that depth, revealed the negative correlation with the surface temperature. The ice sheet elevation in the glacial periods calculated from P_c under the hydrostatic assumption was considerably low, which is quite different from our present knowledge. This means that the total air content in ice is not governed only by atmospheric pressure.

9) $\delta(O_2/N_2)$ from the Dome Fuji core was clearly lower than that in the present atmosphere. By inspecting the storage time of ice samples at high temperature of -25℃ and the correlation of $\delta(O_2/N_2)$ with total air content, it was suggested that faster molecular diffusion of O_2 than N_2 in ice lattice, as well as selective effusion of O_2 at the bottom of firn during the bubble close-off process, caused low values of $\delta(O_2/N_2)$ in ice.

論文審査の結果の要旨

過去の大気組成の変動を再現することは，古気候・古環境を理解する上で不可欠な課題である。川村賢二は，フィルンにおける空気の移動および氷への空気の取り込みの過程を解明した上で，南極ドームふじ基地で掘削された2500mに及ぶ深層氷床コアを高精度で分析し，CH_4，N_2O，CO_2の濃度と大気N_2の$\delta^{15}N$，大気O_2の$\delta^{18}O$，大気中の$\delta(O_2/N_2)$，コア空気含有量の過去340000年にわたる変動を明らかにした。得られた結果は以下のように要約できる。
（1）フィルンの空気は主に分子拡散によって移動することが明らかであり，コア中の各空気成分へ与える影響を，$\delta^{15}N$の分析を基に補正する方法を確立した。また，コアと空気との年代差は，主に堆積環境の変化に伴って氷期－間氷期において2000年から5000年の幅で変化していた。（2）CH_4濃度は，氷期から間氷期へ遷移する際に急増し，間氷期の終わりに急減した後，かなり大きな変動を示しながら，氷期最盛期の最低値へと徐々に減少していた。このような変動は気温と良い相関があり，地球軌道要素の変化に起因する気候変化によって，低緯度から北半球中緯度にかけてのCH_4放出源が影響を受けたためと考えられた。（3）N_2O濃度は，間氷期に高く氷期に低いという変動を示したが，氷期最盛期において非常に高い値がしばしば見いだされ，海面低下に伴って露出された大陸棚での微生物活動がその原因と考えられた。（4）CO_2濃度は氷期－間氷期サイクルとよく同期して190ppmから300ppmの間で変動しており，特に11.5万年の周期の変動が卓越していた。（5）大気の$\delta^{18}O$は，基本的には海面変動を反映して，多少の遅れを伴って海洋の$\delta^{18}O$の変化に追従していたが，間氷期から氷期に転ずる際には関係が逆になっており，陸上生物の寄与が大きいことが示唆された。（6）コア中の$\delta(O_2/N_2)$は大気より明らかに低く，氷への空気取り込み過程においてO_2の選択的逸脱が生じている可能性が指摘された。また，空気含有量は間氷期より氷期に多く，従来の知識とは全く逆の結果となり，新たな支配プロセスの検討を行った。

以上の結果は，川村賢二が自立して研究活動を行うために必要な高度の研究能力と学識を有していることを示している。したがって，川村賢二提出の博士論文は，博士（理学）の学位論文として合格と認める。